Infrared magneto-spectroscopy of graphene-based systems

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S. Winnerl, M. Helm	Helmholtz-Zentrum Dresden, Germany
M. Ostler, Th. Seyller	University of Erlangen-Nuremberg, Germany
M. Polini	NEST, Pisa, Italy
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L. Z. Tan, S. G. Louie	University of California at Berkeley, USA

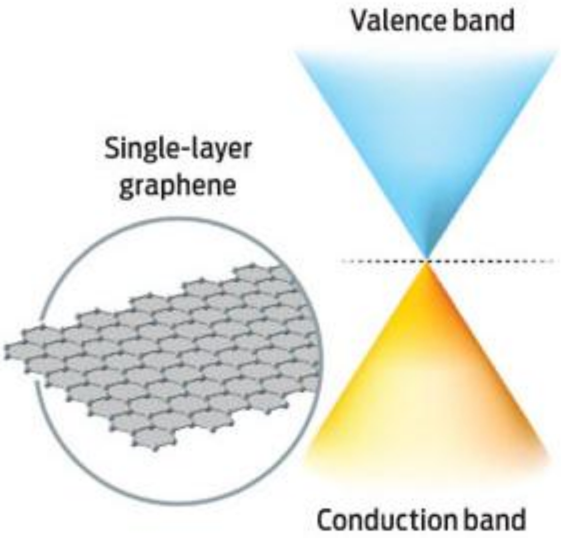
Tainan, 5/07/2012

Outline:

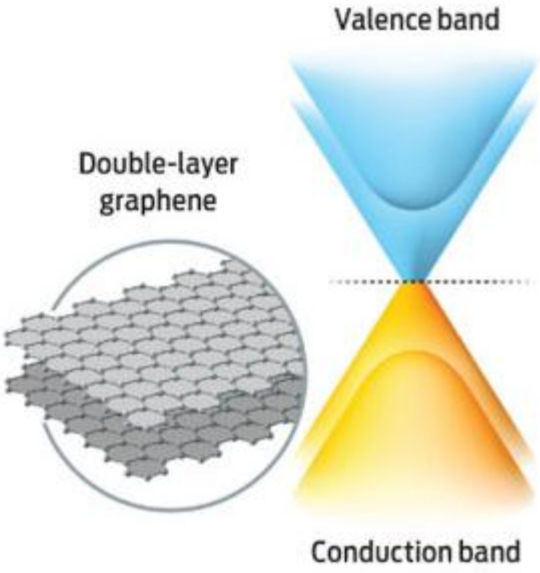
- Introduction into (magneto-)optical response of graphene
- Examples:
    - Cyclotron resonance (CR) in quantum and classical regime
    - Elastic and inelastic scattering of massless Dirac fermions
    - Drude weight versus CR strength (e-e interaction)
    - Confined plasmons and magneto-plasmons
- Conclusions

Physics of graphene-based materials

Graphene:



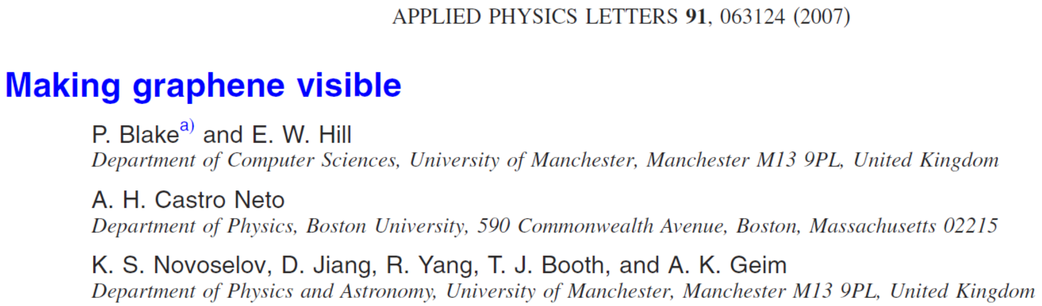
Bilayer graphene:



Credit to IEEE spectrum 2009

Massless and massive Dirac fermions, half-integer quantum Hall effect at room temperature, Berry phase, universal ac conductivity, minimum conductivity, quantum electrodynamics, Klein tunneling, future carbon-based electronics, Nobel prize in Physics 2010, etc.

Optical methods in physics of graphene:
Crucial role from beginning



APPLIED PHYSICS LETTERS **91**, 063124 (2007)

Making graphene visible

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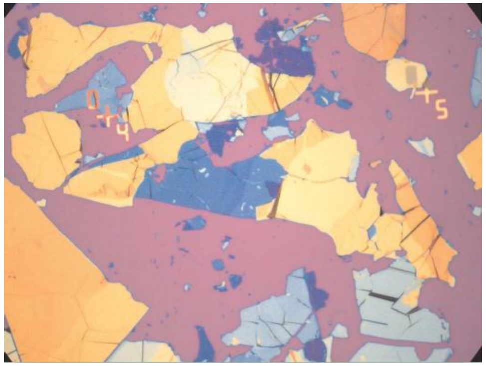
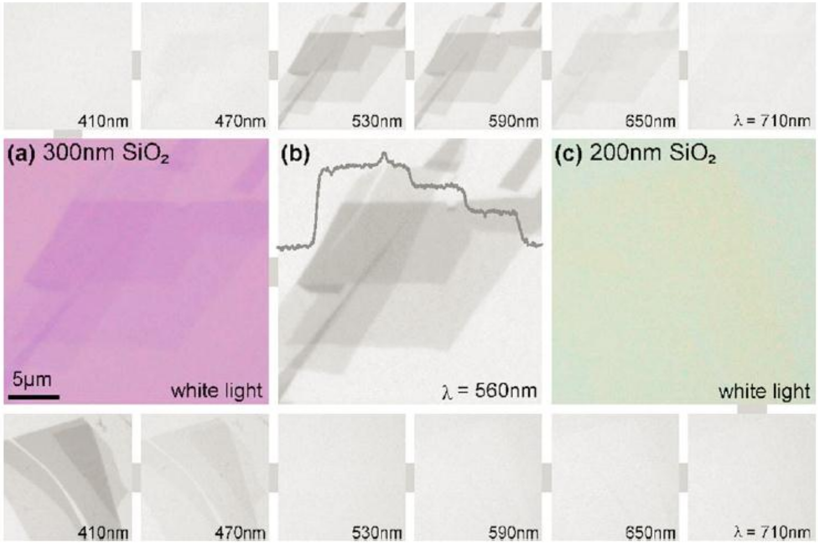
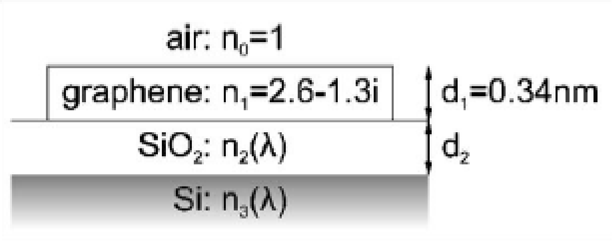
Department of Computer Sciences, University of Manchester, Manchester M13 9PL, United Kingdom

A. H. Castro Neto
Department of Physics, Boston University, 590 Commonwealth Avenue, Boston, Massachusetts 02215

K. S. Novoselov, D. Jiang, R. Yang, T. J. Booth, and A. K. Geim
Department of Physics and Astronomy, University of Manchester, Manchester M13 9PL, United Kingdom







Optical microscopy allowed identification of graphene among other thin graphitic layers on the surface of Si/SiO$_2$

Optical probing of graphene

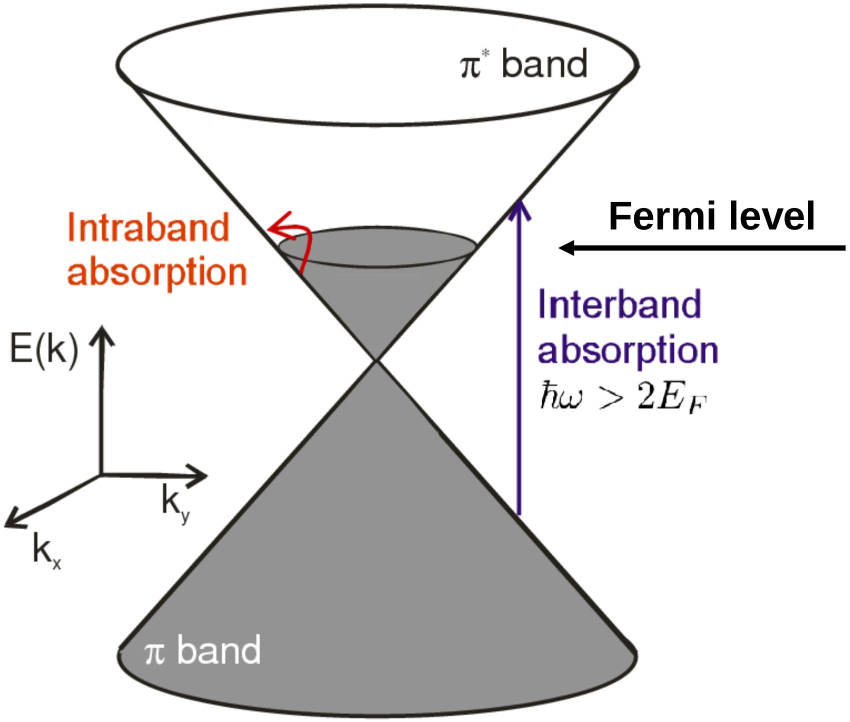
Intraband (Drude, free carrier) absorption

- Sensitive to the vicinity of the Fermi level

Interband absorption

- Absorption onset at $2E_F$

- Probe of occupied and empty states away from the Fermi level



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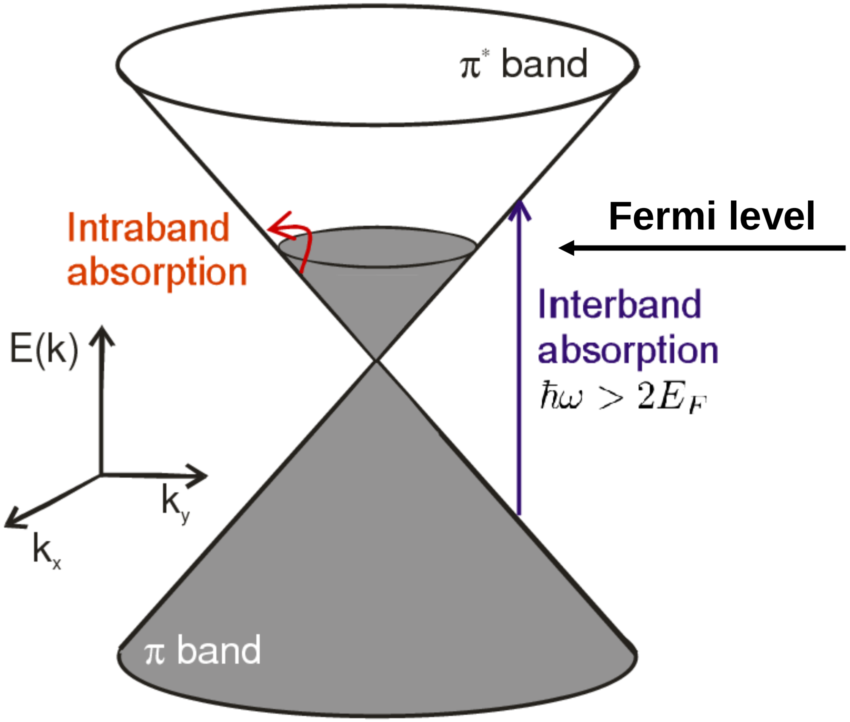
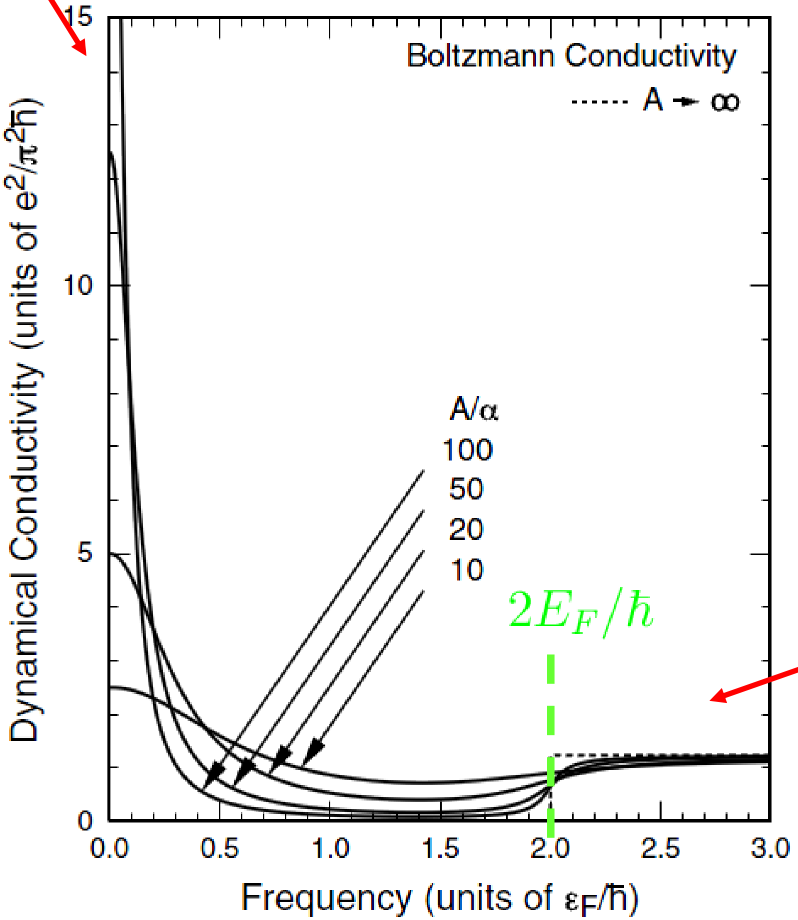


How to extract information on electronic band structure, structural defects, type of dominant scatterers, carrier relaxation time, electron-electron interaction, electron-phonon interaction….

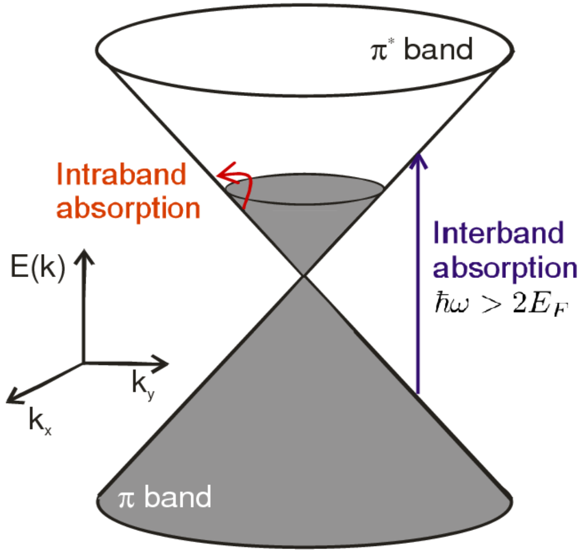
Dynamical conductivity: Theory

Drude-type absorption (intraband)

Sensitive to the vicinity of the Fermi level



T. Ando, J. Phys. Soc. Jpn. 71, 1318 (2002)



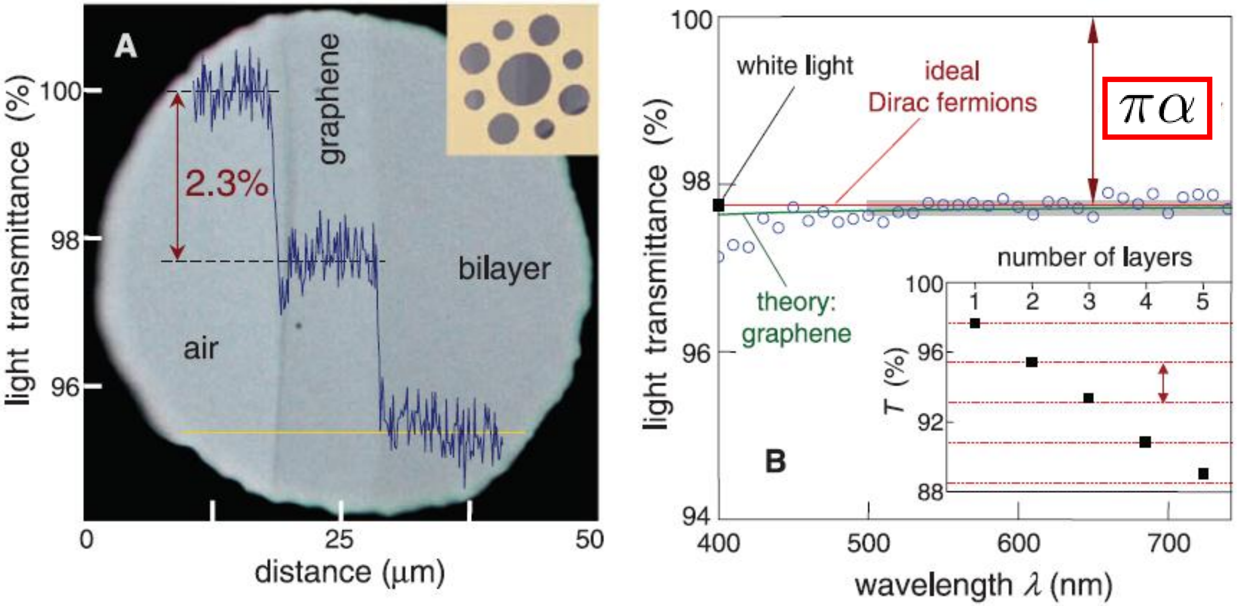
Interband transitions

Probe of occupied and empty states away from the Fermi level

Flat for $\hbar\omega \gg 2E_F$

R. R. Nair et al., Science 320, 1308 (2008)

Universal ac conductivity of graphene



Interband absorption in graphene is flat (2.3%) and depends on the fine structure constant only

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R. R. Nair et al., Science 320, 1308 (2008); A. B. Kuzmenko et al., PRL 100, 17401 (2008)

Dynamical conductivity: Theory

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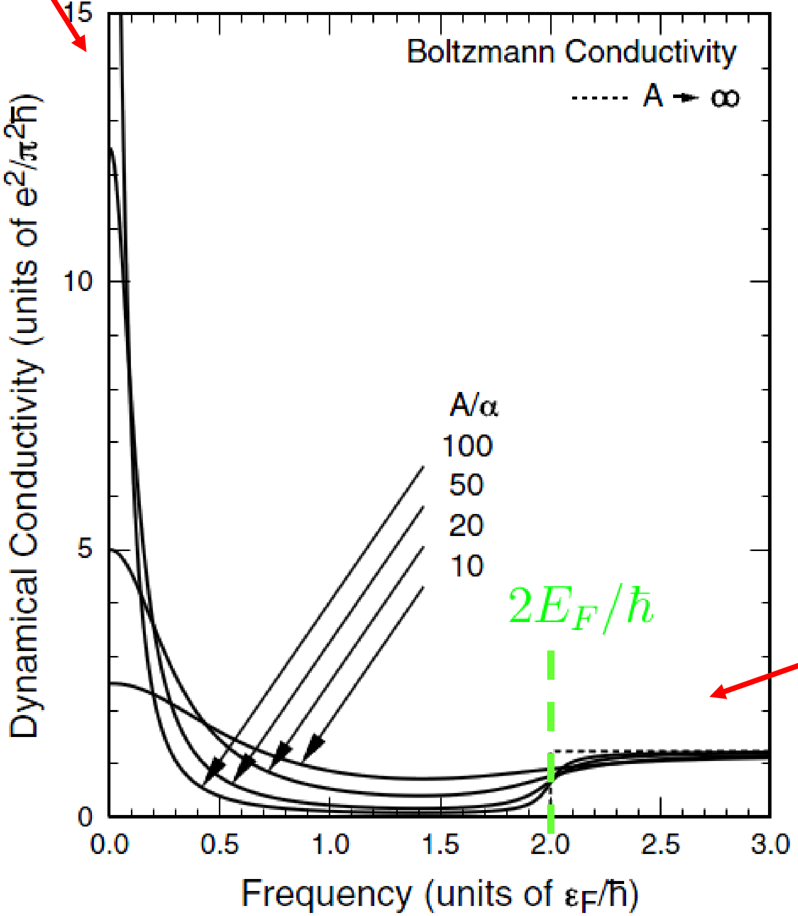
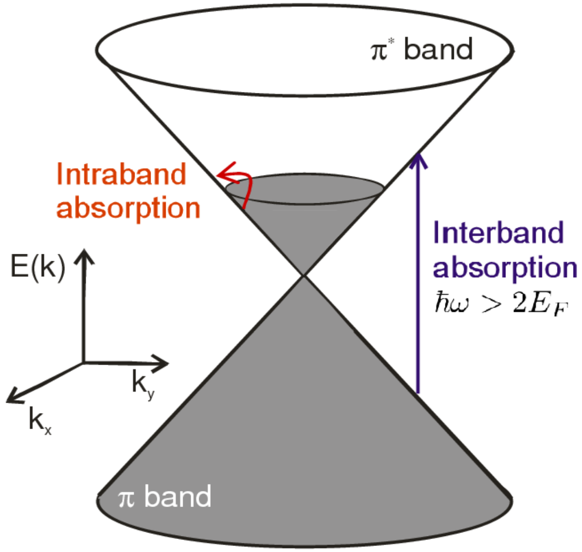
Sensitive to the vicinity of the Fermi level





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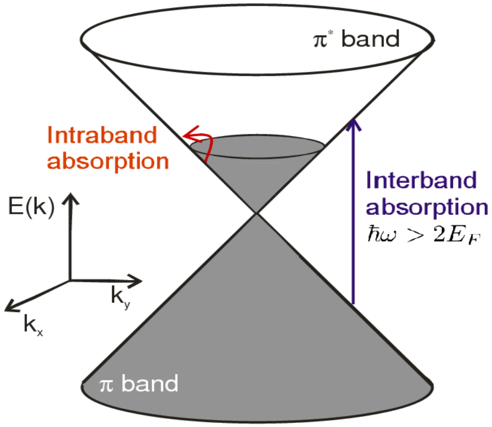
R. R. Nair et al., Science 320, 1308 (2008)

T. Ando, J. Phys. Soc. Jpn. 71, 1318 (2002)

Dynamical conductivity at $B \neq 0$



Drude-type absorption (intraband) ⇒ **Cyclotron resonance (CR)**

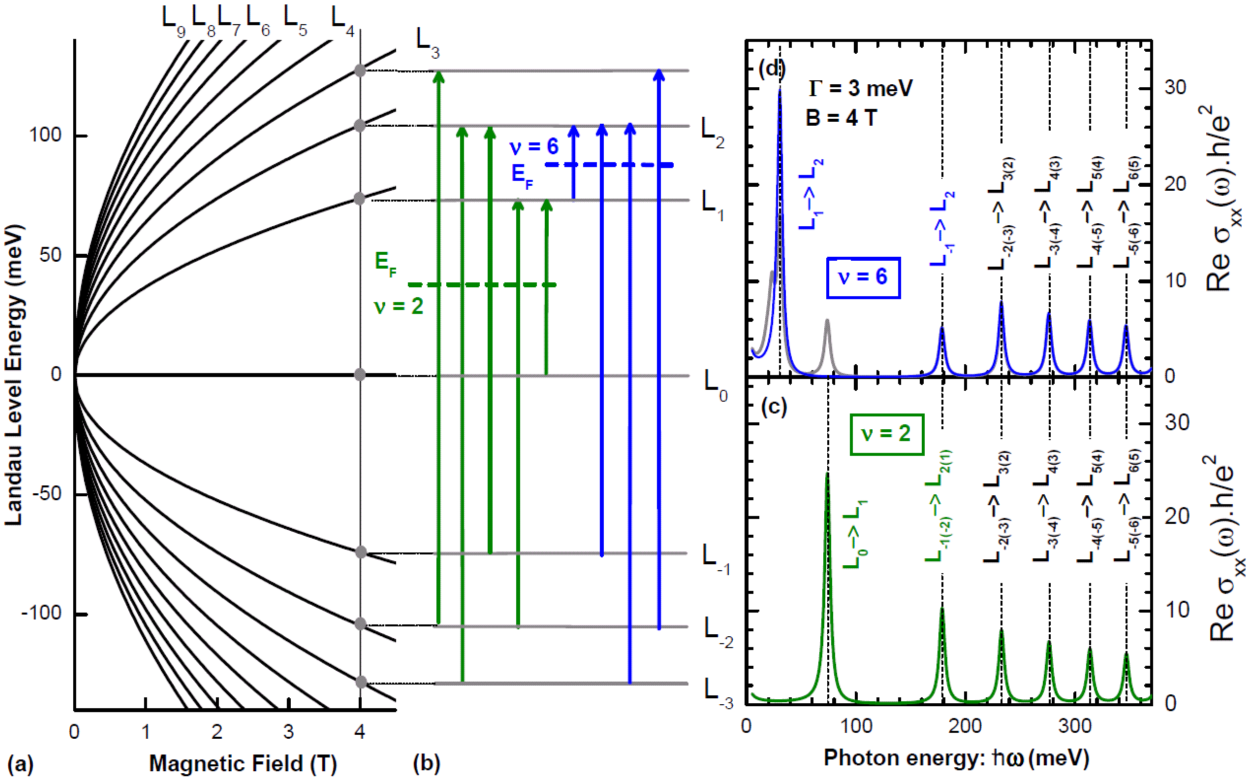
(Flat) interband absorption ⇒ **Interband inter–Landau level transitions**

Energy spectrum:

$$E_n = \pm v_F \sqrt{2e\hbar|Bn|}$$

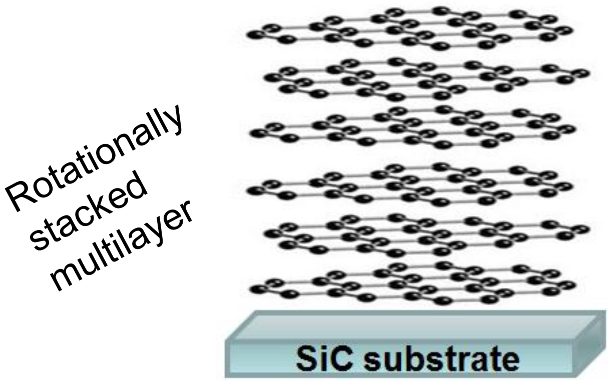
Selection rules:

$$|n| \rightarrow |n| \pm 1$$



Samples and experimental technique

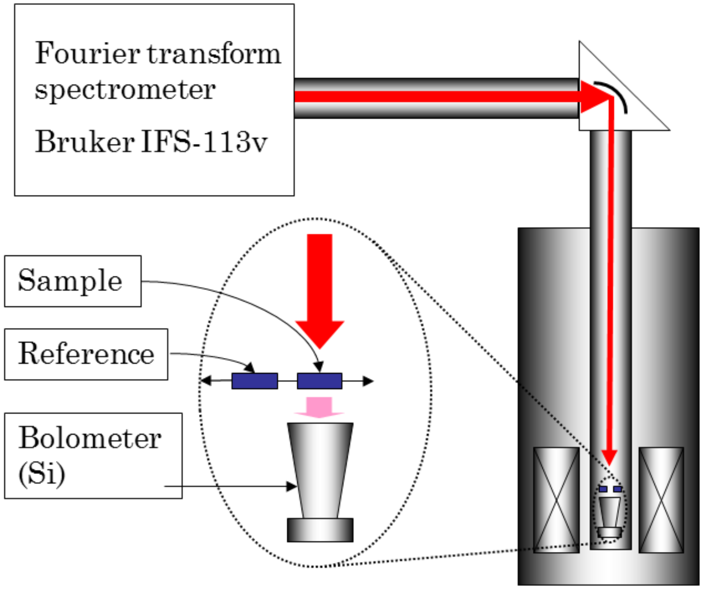
Multilayer epitaxial graphene on C-face of SiC (MEG):



High-quality, quasi-neutral layers
Nice for optics, complex for transport…

M. Sadowski et al., Phys. Rev. Lett. 97, 266405 (2006)
J. Hass et al, Phys. Rev. Lett. 100, 125504 (2008)

Far infrared magneto-spectroscopy (transmission configuration):

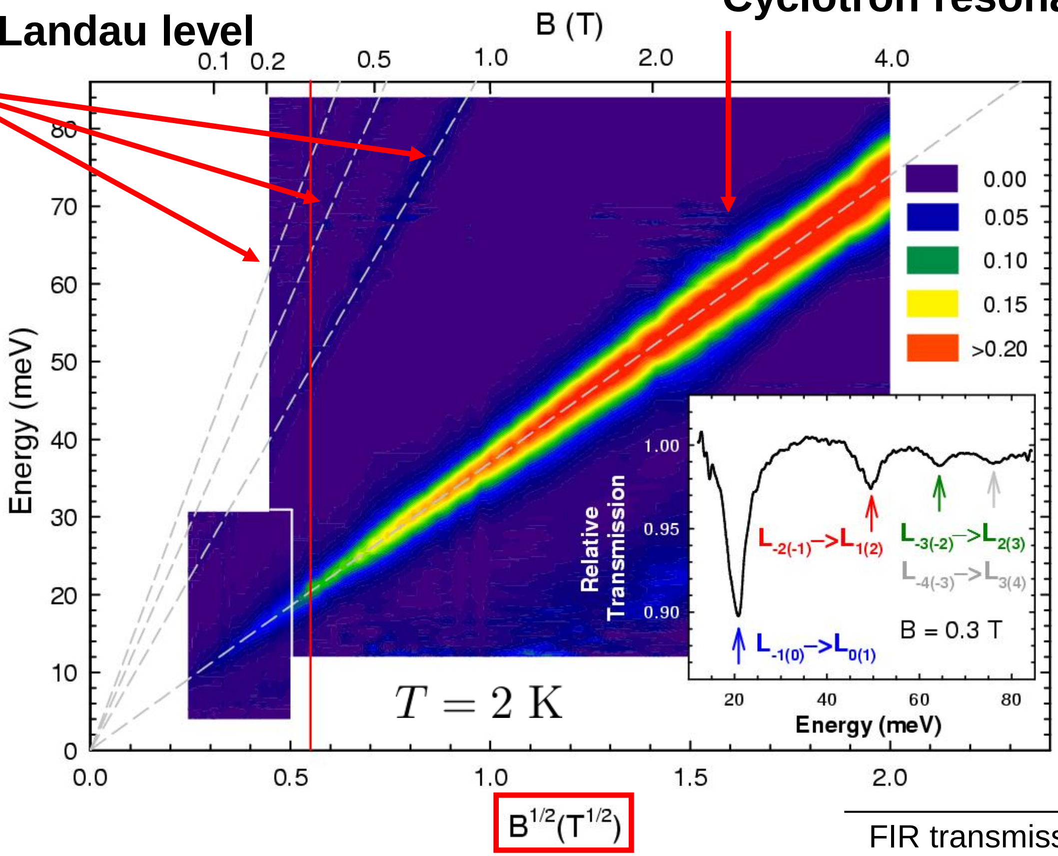


Samples: GeorgiaTech – Atlanta, ITME – Warsaw, Linköping University

Magneto-transmission of (multilayer epitaxial) graphene

Interband inter-Landau level transitions

Cyclotron resonance



FIR transmission *T* plotted as –*ln(T)*

Energy spectrum:

$$E_n = \pm v_F \sqrt{2e\hbar|Bn|}$$

Selection rules:

$$|n| \rightarrow |n| \pm 1$$

M. L. Sadowski et al., PRL 97, 266405 (2006)
Z. Jiang et al., PRL 98, 197403 (2007)
M. Orlita et al., PRL 101, 267601 (2008)

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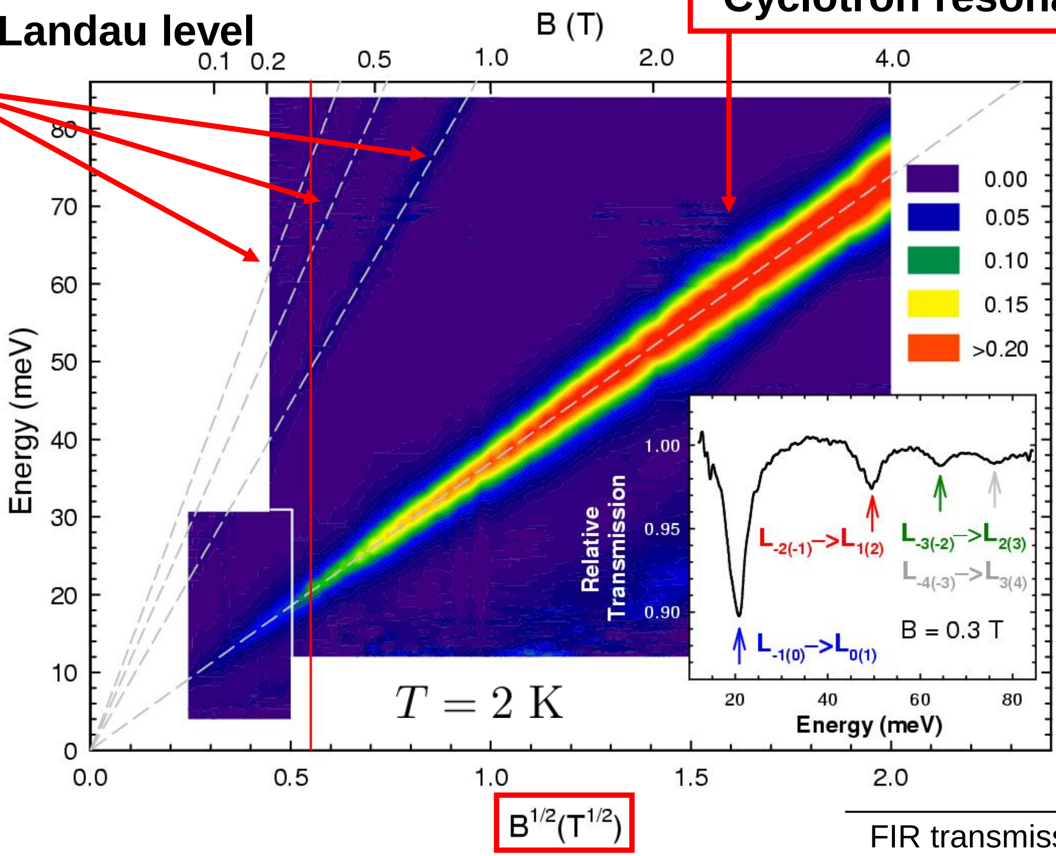

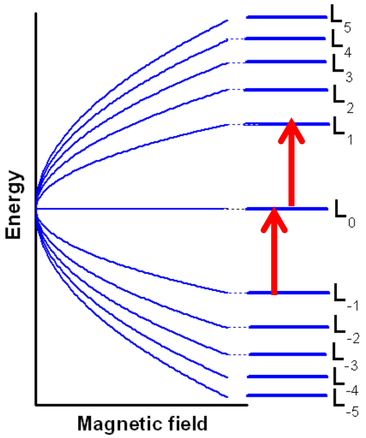

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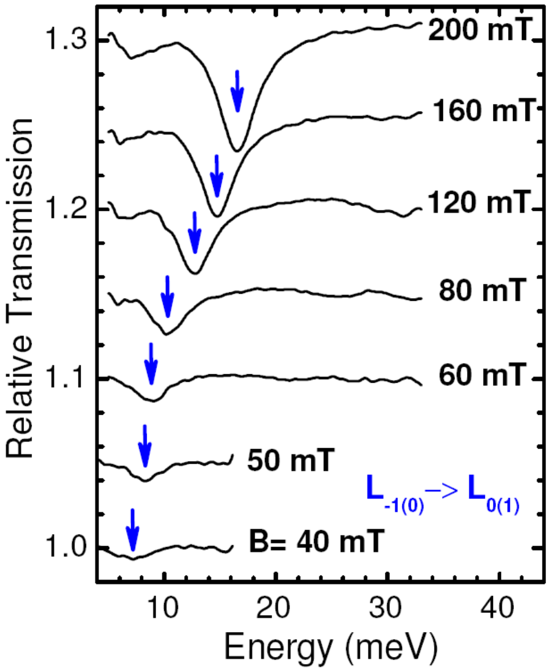
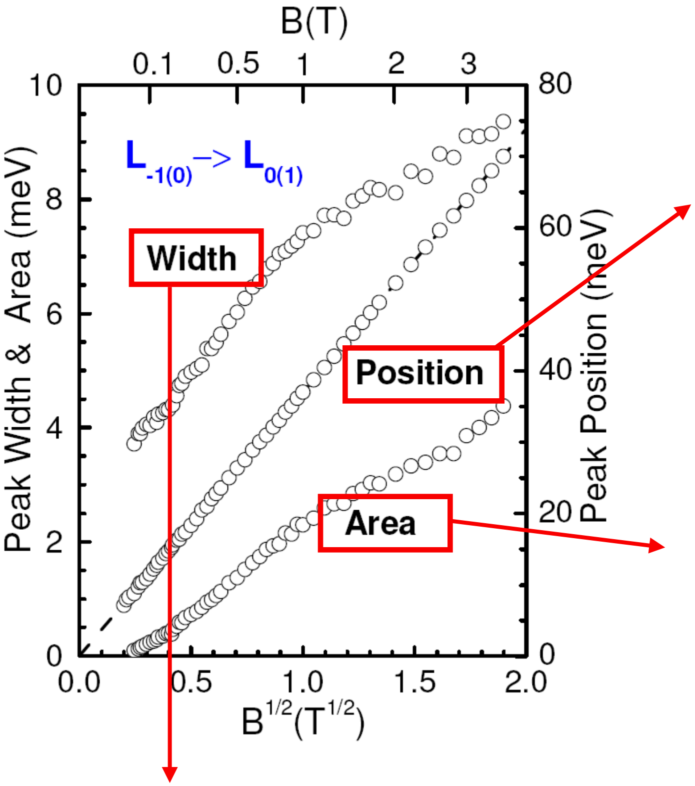
M. L. Sadowski et al., PRL 97, 266405 (2006)
Z. Jiang et al., PRL 98, 197403 (2007)
M. Orlita et al., PRL 101, 267601 (2008)

Analysis of CR response





Fermi velocity
(from CR energy)

$$v_F = 1.02 \times 10^6 \text{ m/s}$$

Carrier density
(from CR intensity, $\nu \approx 6$)

$$n \approx 5 \times 10^9 \text{ cm}^{-2}$$

Scattering time
(from CR width)

$$\tau(E_F) \sim 300 \text{ fs}$$

Carrier mobility?

$$\tau\omega_c > 1 \Rightarrow \mu B > 1$$

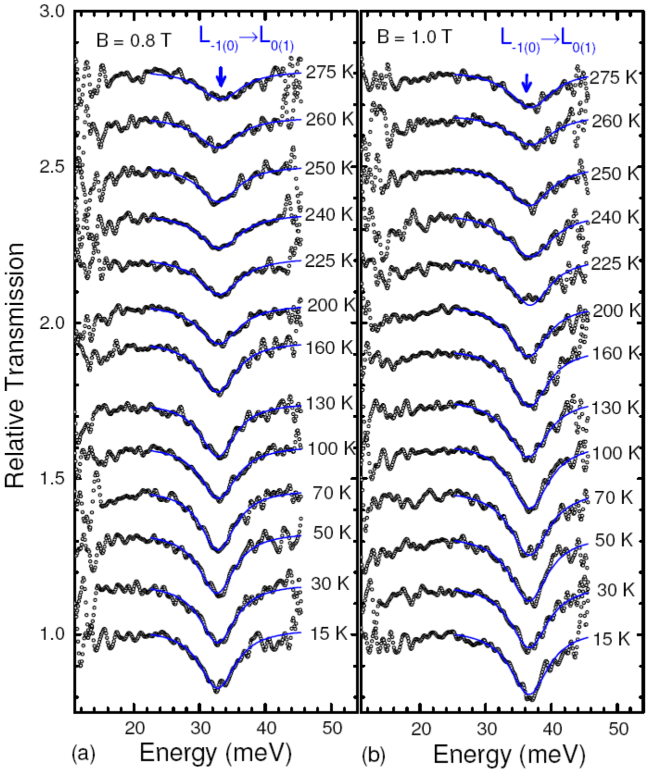
Main line down to $B = 40 \text{ mT}$

⇨

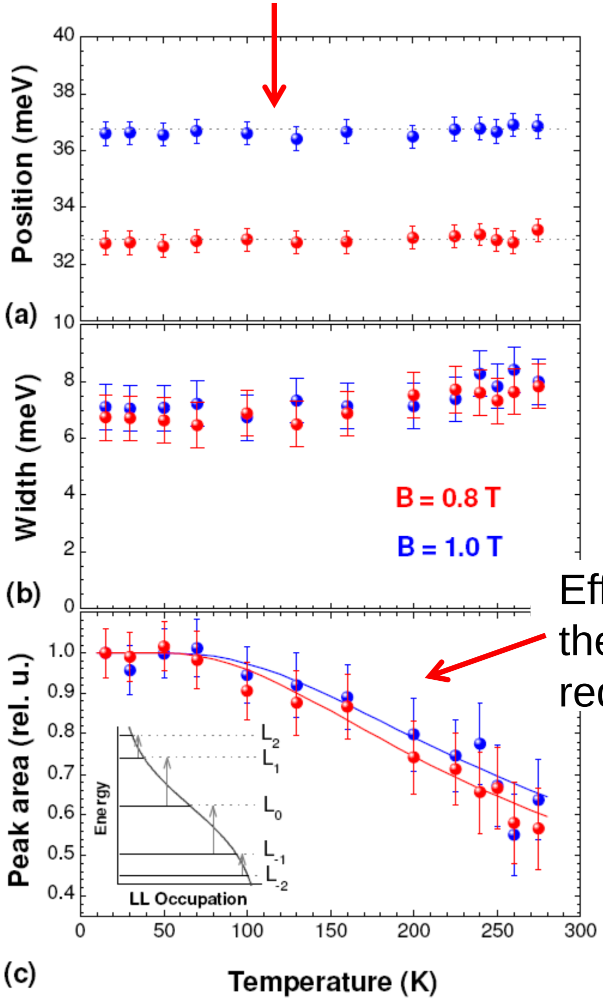
$$\boxed{\mu > 250000 \text{ cm}^2/(\text{V.s})}$$

M. Orlita et al., PRL 101, 267601 (2008)

Temperature dependence



Fermi velocity independent of temperature



Effect of thermal redistribution

No scattering process activated up to RT.
Record mobility at RT?

$$\mu_{\text{Graphene}} > 250\,000\ \text{cm}^2/(\text{V.s})$$

M. Orlita et al., PRL 101, 267601 (2008)

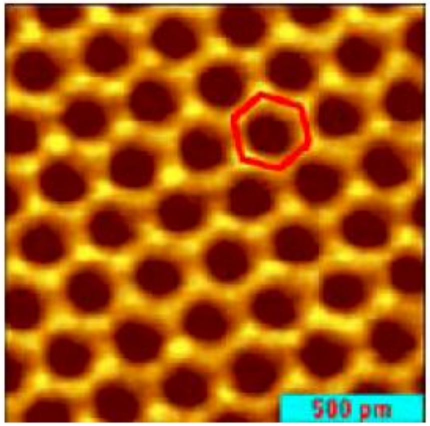
Comparison of RT mobilities:

$$\mu_{\text{Si}} \approx 1\,500\ \text{cm}^2/(\text{V.s}) \qquad \mu_{\text{GaAs}} \approx 8\,500\ \text{cm}^2/(\text{V.s}) \qquad \mu_{\text{InSb}} \approx 77\,000\ \text{cm}^2/(\text{V.s})$$

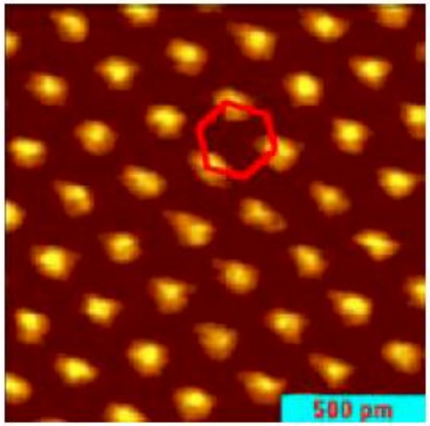
Cyclotron resonance in graphene on graphite

Graphene flakes on graphite:

G. Li et al., PRL 102, 176804 (2009)

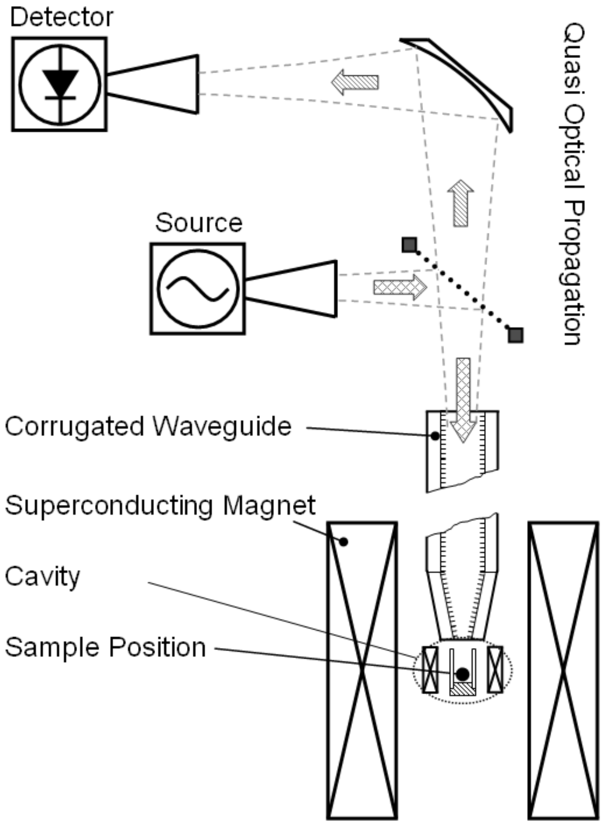


Weakly doped graphene sheets



AB stacked bulk graphite

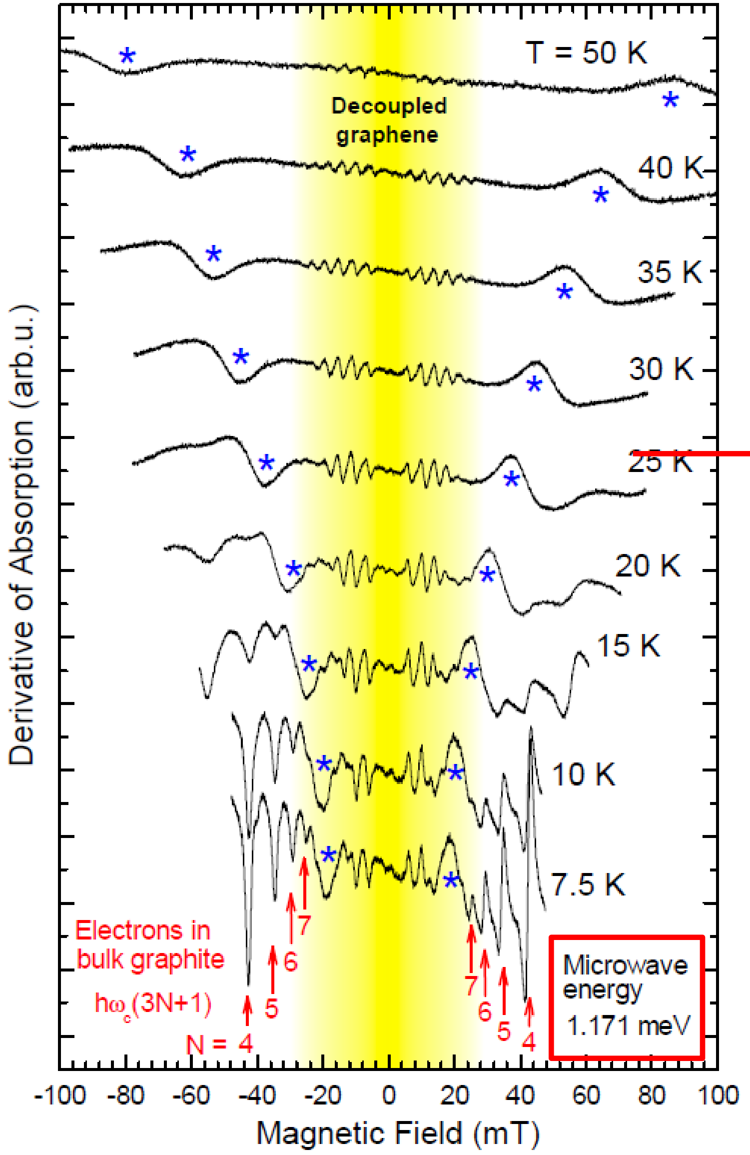
Landau level spectroscopy using EPR setup (THz range):



Field-modulation technique employed

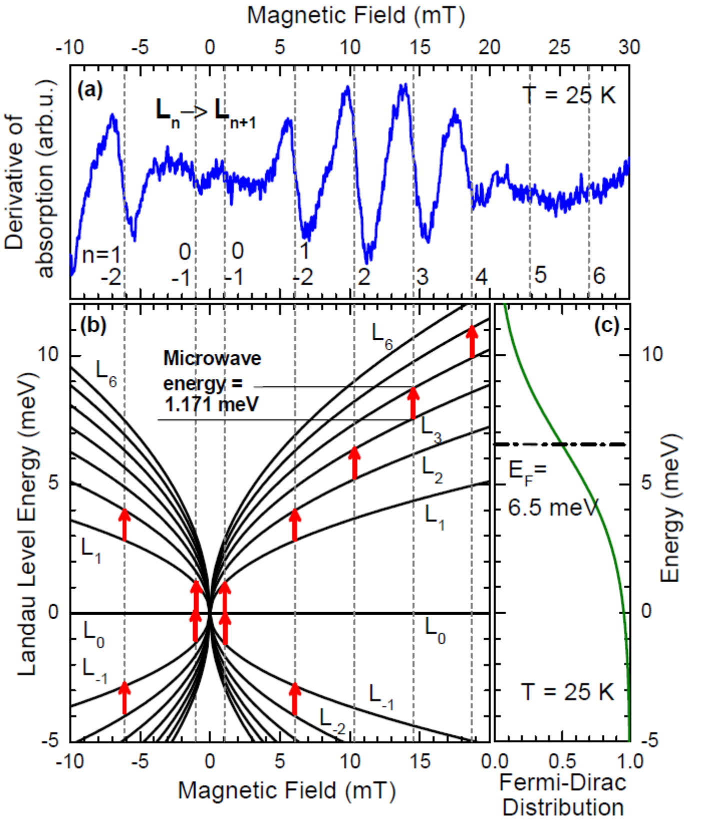
Cyclotron resonance in graphene on graphite

CR in sub-THz range:



Energy spectrum: $E_n = \pm v_F \sqrt{2e\hbar|Bn|}$

Non-equidistant LL spacing $\lesssim kT$ ⇨ **Multimode CR**



P. Neugebauer, M. Orlita, et al., PRL 103, 136403 (2009)

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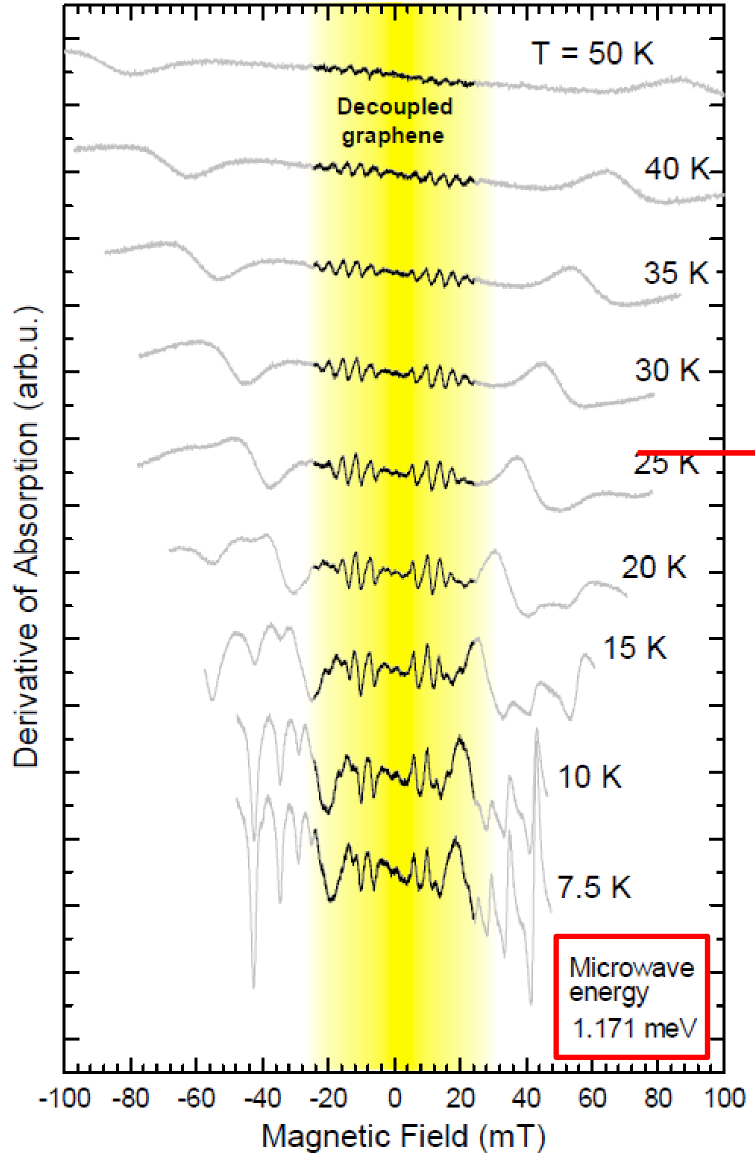
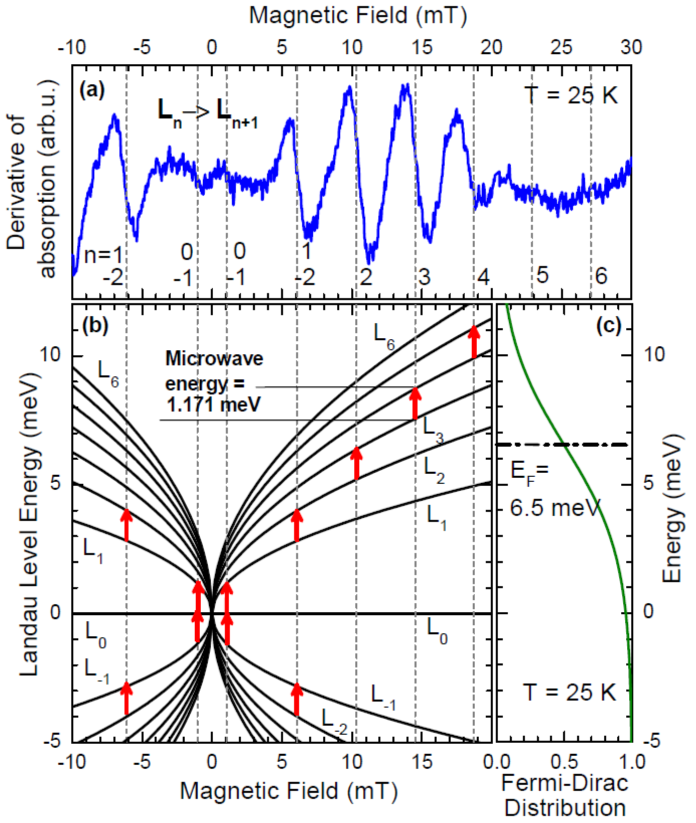




P. Neugebauer, M. Orlita, et al., PRL 103, 136403 (2009)

Analysis of (multimode) CR in graphene on graphite

Fermi velocity:

$$v_F = (1.00 \pm 0.02) \times 10^6 \text{ m/s}$$

Fermi level & density:

$$E_F \approx 6 \text{ meV}$$

$$n \approx 3 \times 10^9 \text{ cm}^{-2}$$

Resonance widths: $\delta E \approx 50\ \mu\text{eV}$

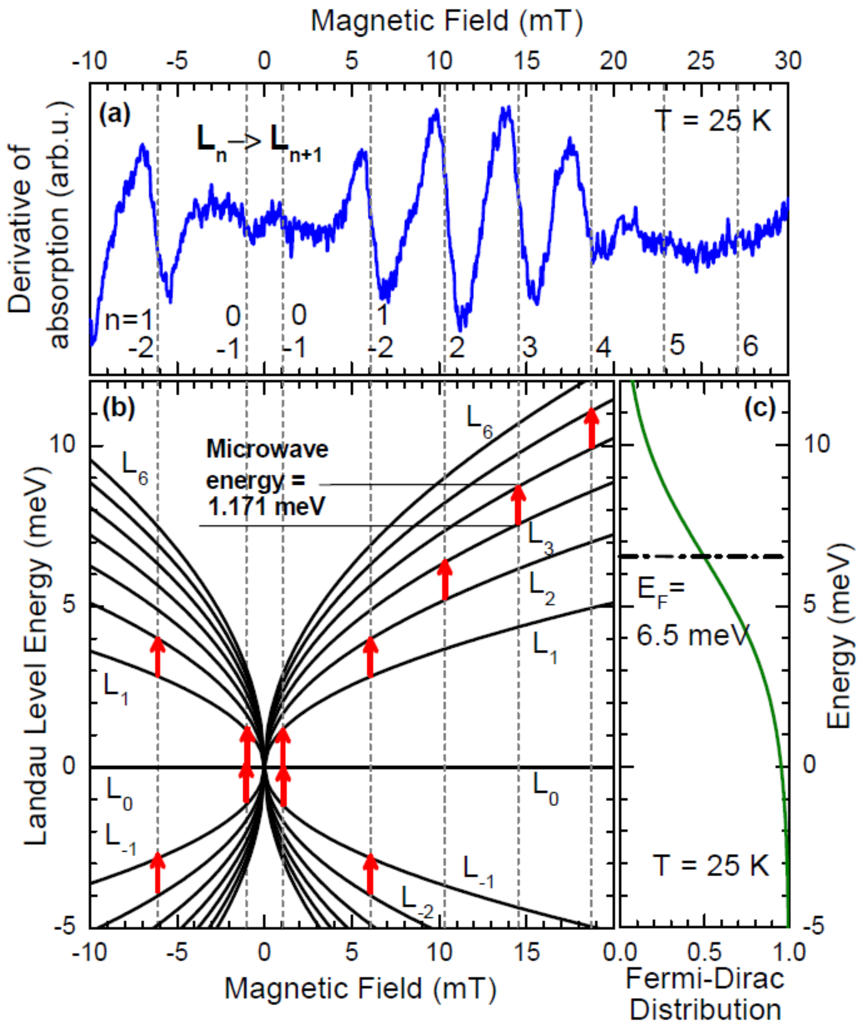
$$\Rightarrow \tau(E_F) \approx 20 \text{ ps}$$

LL quantization down to $B = 1$ mT

⇩

High-mobility graphene!

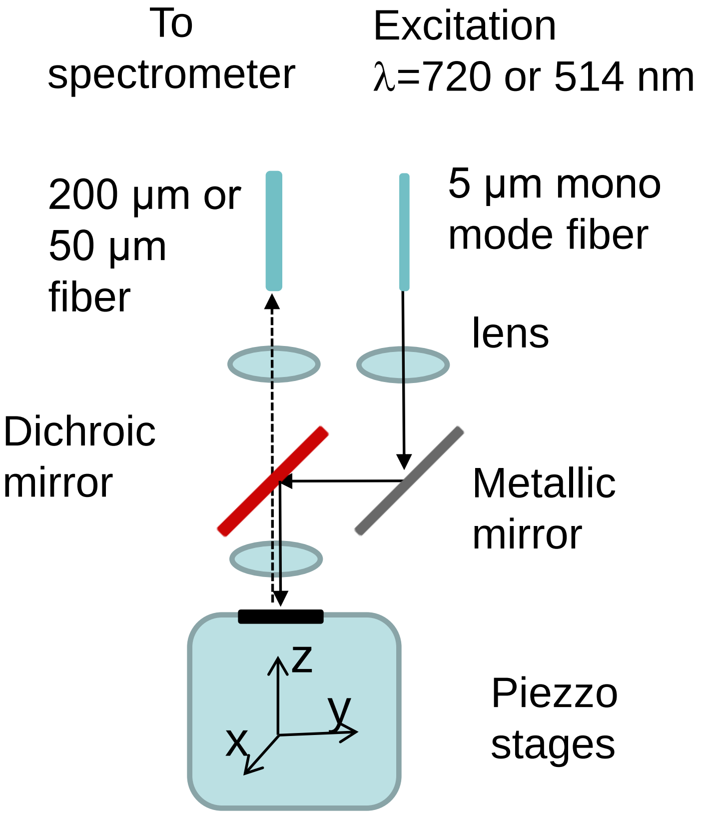
$$\mu > 10 \times 10^6 \text{ cm}^2/(\text{V.s})$$



P. Neugebauer, M. Orlita, et al., PRL 103, 136403 (2009)

Mapping surface of natural graphite

Micro-Raman in high magnetic fields:



To spectrometer

Excitation λ=720 or 514 nm

200 µm or 50 µm fiber

5 µm mono mode fiber

lens

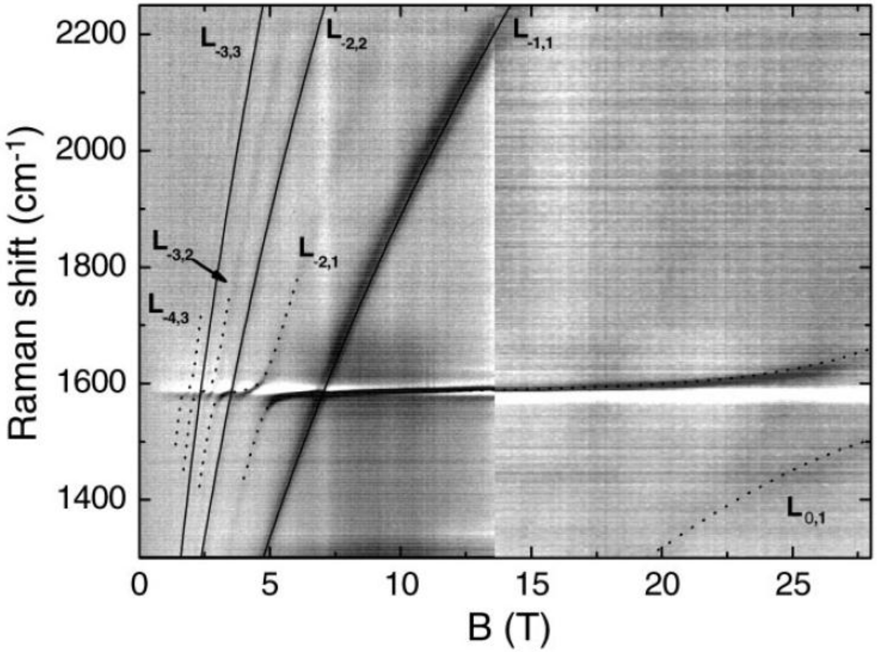
Dichroic mirror

Metallic mirror

Piezzo stages

T = 4.2 K, P ~ 5 mW, fields up to 30 T
Spot size~ 1µm
Resolved to circular polarization

Magneto-Raman spectra from graphene on graphite:



Inter-Landau level excitations

(The first purely electronic Raman excitation in graphene-like systems)

C. Faugeras et al., PRL 107, 036807 (2011)

Magneto-phonon effect

(Raman non-active excitations coupled to E_{2g} phonon)

C. Faugeras et al., PRL 103, 186803, (2009)

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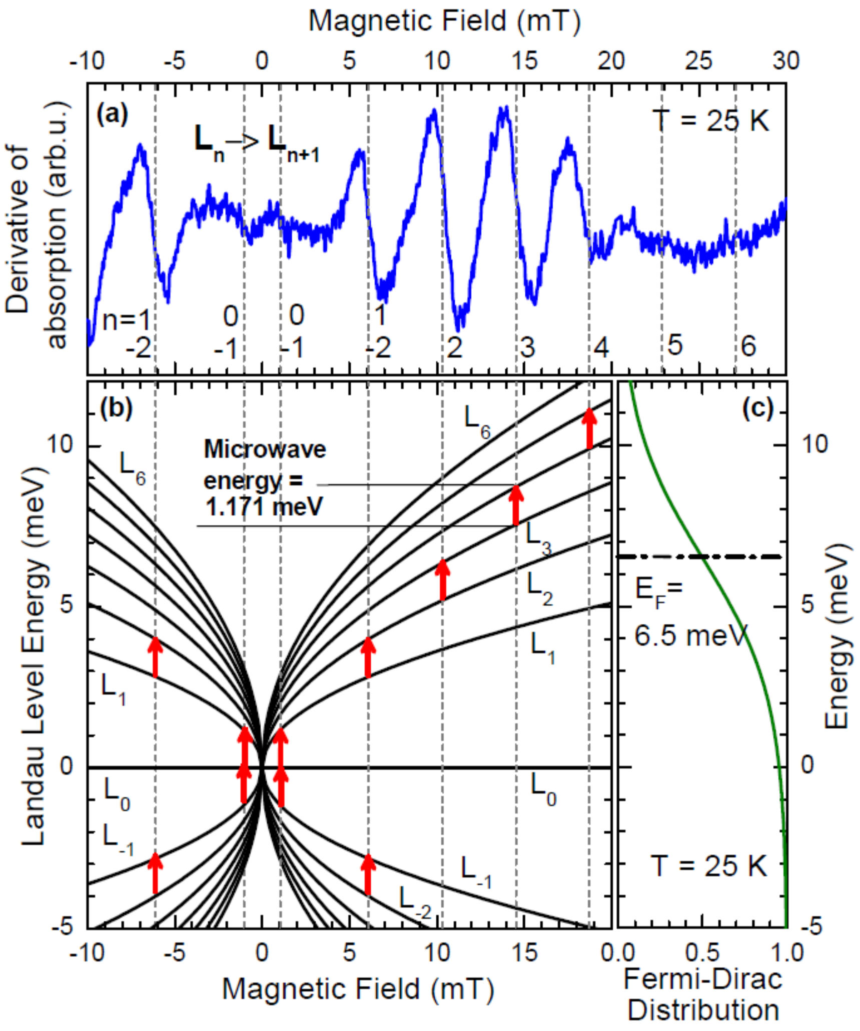


P. Neugebauer, M. Orlita, et al., PRL 103, 136403 (2009)

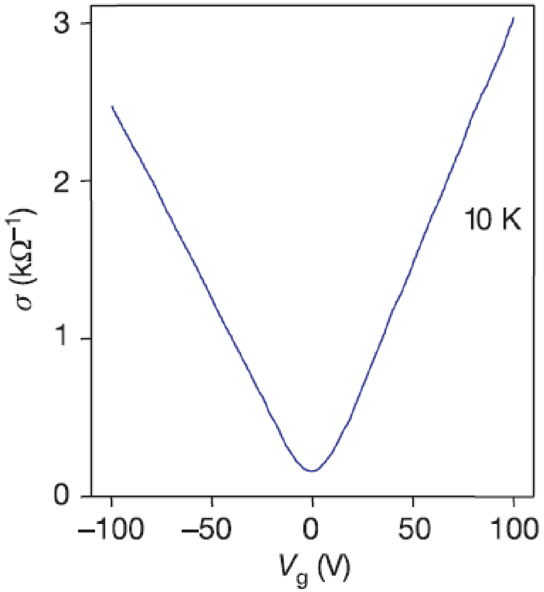
Relaxation time & dc conductivity in graphene

Experiments (on gated flakes):

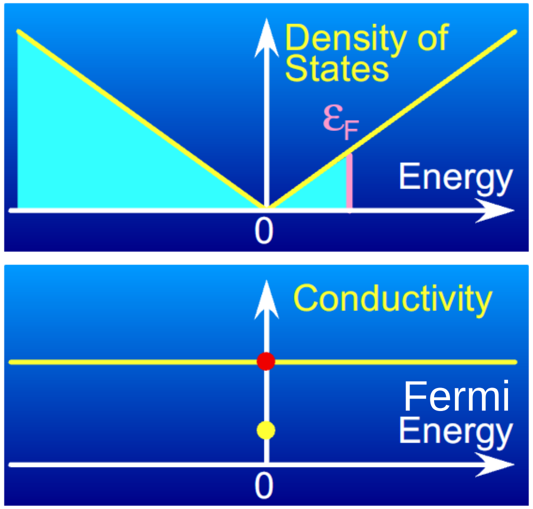
$$\boxed{\sigma_{dc} = e\mu n}$$

$$\mu = \text{const} = e^2 \frac{\tau(E_F)}{m} = e^2 v_F^2 \frac{\tau(E_F)}{E_F}$$

⇨ $\tau(E_F) \propto |E_F|$



K. S Novoselov et al., Nature 428, 197 (2005)
Y. Zhang et al., Nature 428, 201 (2005)



T. Ando, J. Phys. Soc. Jpn. 71, 1318 (2002)

Boltzmann conductivity: $\sigma_{dc} \propto v_F^2 \cdot \tau \cdot D$

Density of states: $\mathcal{D}(E) \propto |E|$

Relaxation time
(for “common” scatterers): $\tau(E) \propto |E|^{\boxed{-1}}$

⇨ $\boxed{\sigma_{dc} = \text{const}}$

Explanation?

How to get scattering time increasing with Fermi level…

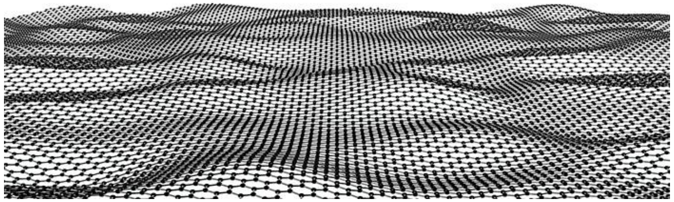
$$\tau(E_F) \propto |E_F|$$

Charged impurities

T. Ando, J. Phys. Soc. Jpn. 75, 074716 (2006)
K. Nomura and A. H. MacDonald, PRL 96, 256602 (2006)
E. H. Hwang, S. Adam, and S. Das Sarma, PRL 98, 186806 (2007)
L. A. Ponomarenko et al., PRL 102, 206603 (2009)

Special type of rippling

M. I. Katsnelson and A. K. Geim,
Phil. Trans. R. Soc. A 366, 195 (2008)



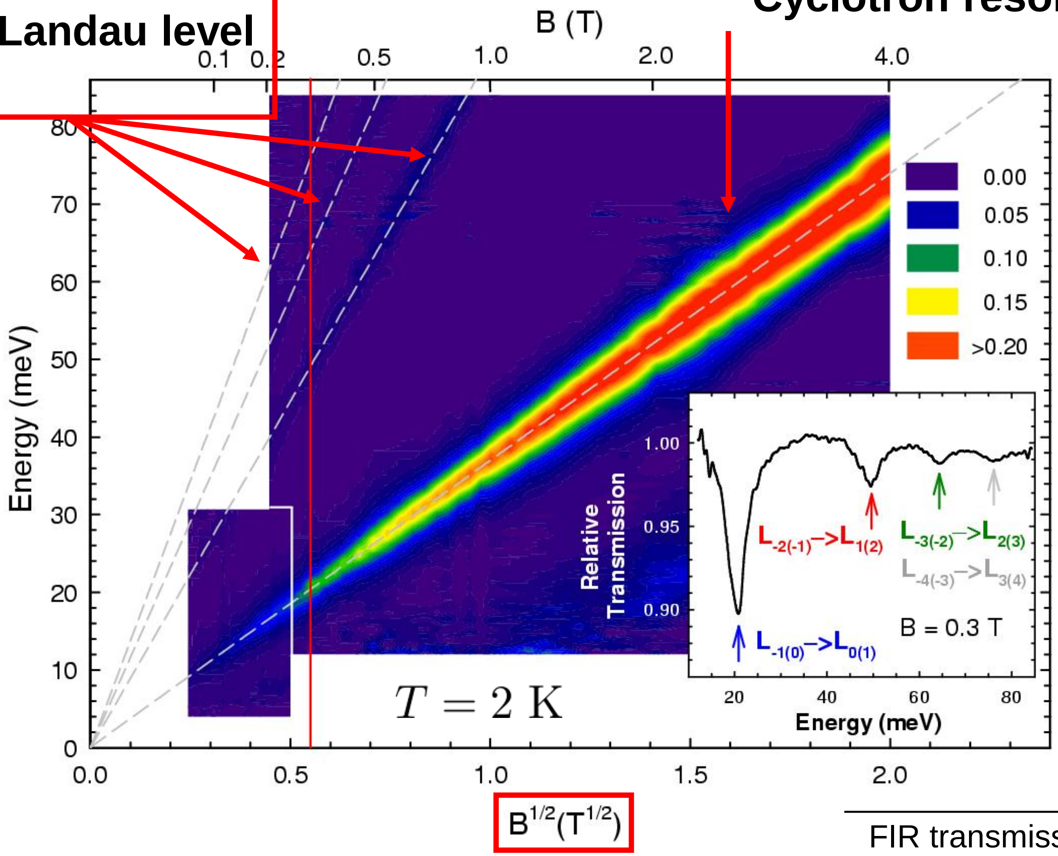
Resonant scatterers (midgap states)

Z. H. Ni et al., Nano Letters 10, 3868 (2010)
T. O. Wehling et al., PRL 105, 056802 (2010)

Magneto-transmission of (multilayer epitaxial) graphene

Interband inter-Landau level transitions

Cyclotron resonance



FIR transmission *T* plotted as *–ln(T)*

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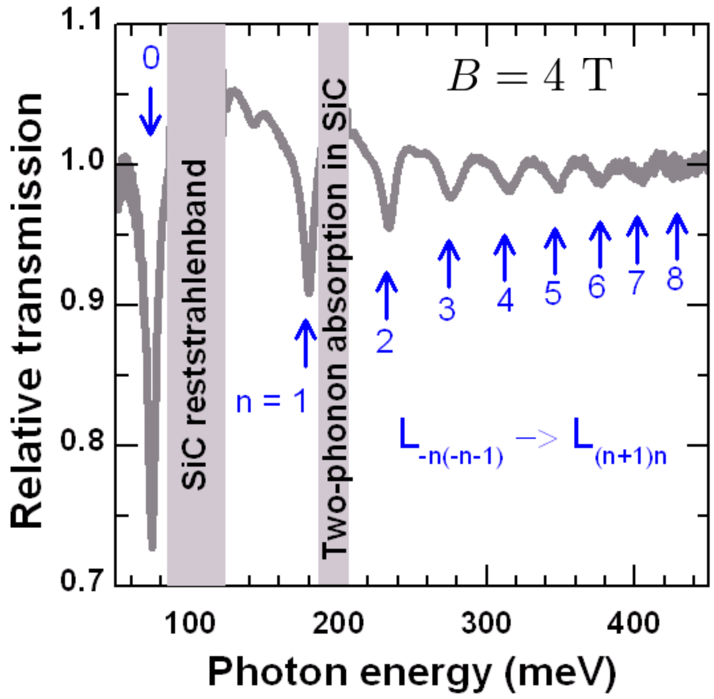
M. L. Sadowski et al., PRL 97, 266405 (2006)
Z. Jiang et al., PRL 98, 197403 (2007)
M. Orlita et al., PRL 101, 267601 (2008)

Dynamical conductivity: quantitative analysis



How to extract energy dependence of relaxation time? $\Rightarrow \tau(E)$

Dynamical magneto-conductivity (Kubo-Greenwood):

$$\sigma_{xx}(\omega, B) \propto \frac{B}{\omega} \sum_{m,n} M_{m,n} \frac{f_n - f_m}{E_m - E_n - (\hbar\omega + \boxed{i\Gamma})}$$

$$\Downarrow$$

Phenomenological broadening:

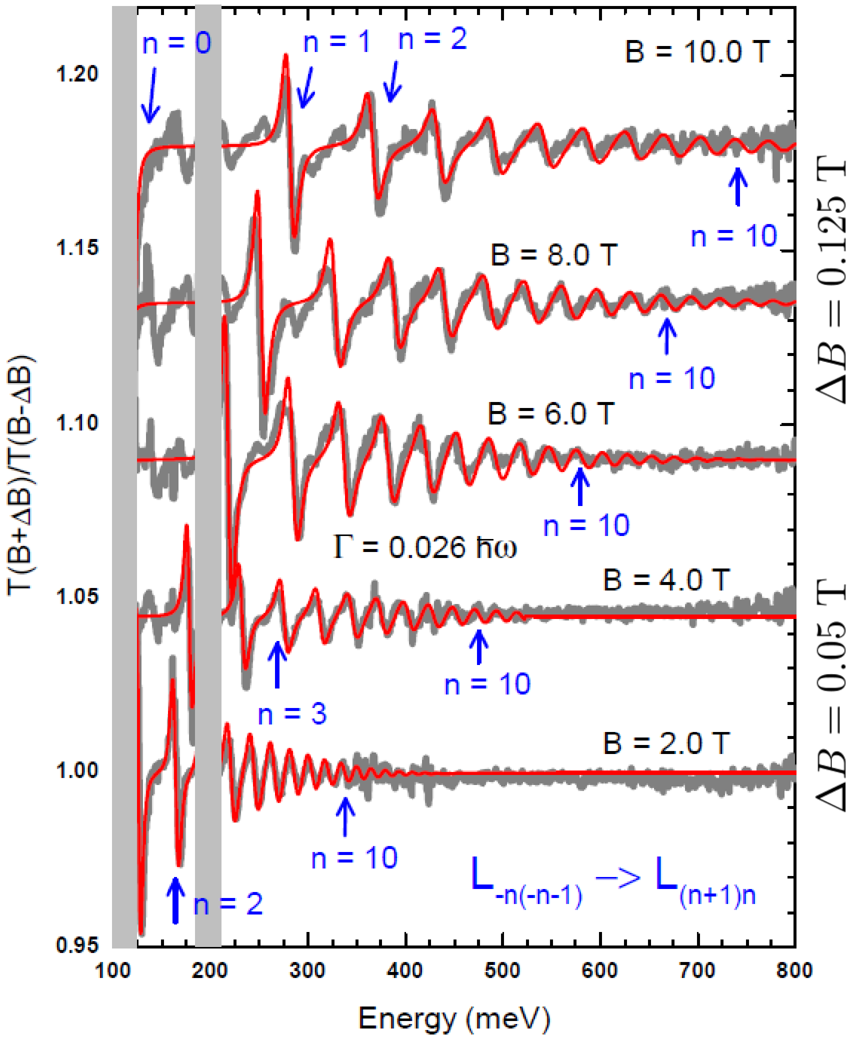
Γ …width of n-th LL (for well separated levels)

$$\boxed{\Gamma \sim \hbar/\tau(E)}$$

…relaxation time
(for strongly overlapping levels)

Landau quantization in epitaxial graphene

Differential spectra (…high energy part):



Fixed number of LLs resolved

LLs resolved up to energy

$$\epsilon \propto \sqrt{B}$$

Onset of LL quantization

$$\omega_c \tau \sim 1$$

Cyclotron frequency:

$$\omega_c = v_F^2 eB/\epsilon$$

⇩

Relaxation time:

$$\tau(E) \propto |E|^{-1}$$

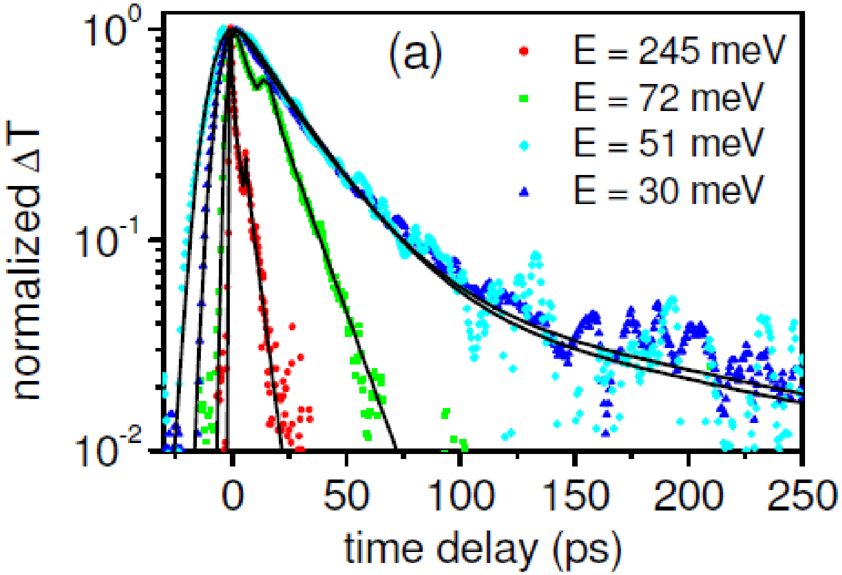
M. Orlita et al., PRL 107, 216603 (2011)

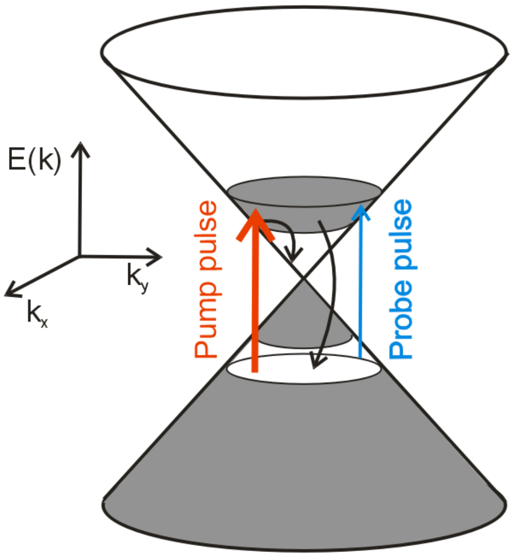
Relaxation dynamics –
Pump and probe experiments

FELBE

HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

S. Winnerl & M. Helm





E(meV)	τ_{pulse} (ps)	τ_1 (ps)	τ_2 (ps)	τ_3 (ps)
245	0.7	0.5 ± 0.1	5.2 ± 0.2	within noise
72	3	-	14.5 ± 1	300 ± 50
51	11	-	25 ± 2	300 ± 50
30	7	-	25 ± 2	300 ± 50

Inelastic scattering more than order of magnitude slower

S. Winnerl, M. Orlita et al., PRL 107, 237401 (2011)

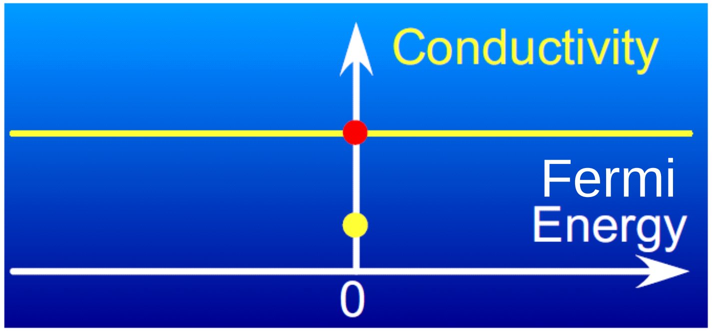
Dynamical versus dc conductivity: Discussion

Relaxation time

$$\tau(E) \propto |E|^{-1}$$

In agreement with original expectations for "2D graphite"

T. Ando, J. Phys. Soc. Jpn. 71, 1318 (2002)



dc conductivity of epitaxial graphene?

C. Berger et al., J. Phys. Chem. B 108, 19912 (2004)
X. Wu et al., Appl. Phys. Lett. 95, 223108 (2009)
K.V. Emtsev et al., Nature Mater. 8, 203 (2009)
Yu-Ming Lin et al., Appl. Phys. Lett. 97, 112107 (2010) etc.

Multilayer C-face (not at interface):

$$n \lesssim 10^{10}\ \mathrm{cm}^{-2}, \mu \gtrsim 10^{5}\ \mathrm{cm}^2/\mathrm{V.s}$$

Monolayer C-face:

$$n \sim 10^{11}\mathrm{cm}^{-2}, \mu \sim 10^{4}\ \mathrm{cm}^2/\mathrm{V.s}$$

Monolayer Si-face:

$$n \gtrsim 10^{12}\ \mathrm{cm}^{-2}, \mu \sim 10^{3}\ \mathrm{cm}^2/\mathrm{V.s}$$

Surprisingly similar conductivities in very different specimens…

$$\sigma_{dc} \sim 10^{-3}\Omega^{-1} \approx 100e^2/h$$

Dynamical versus dc conductivity: Discussion

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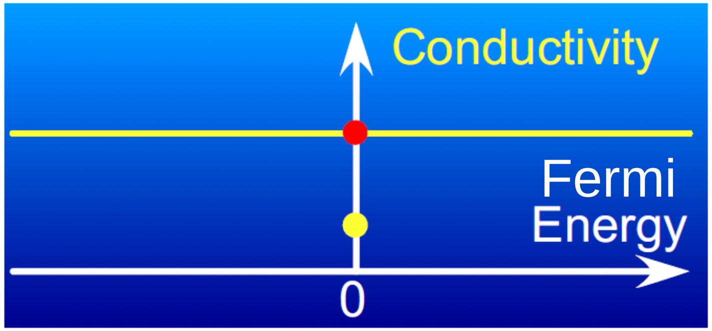

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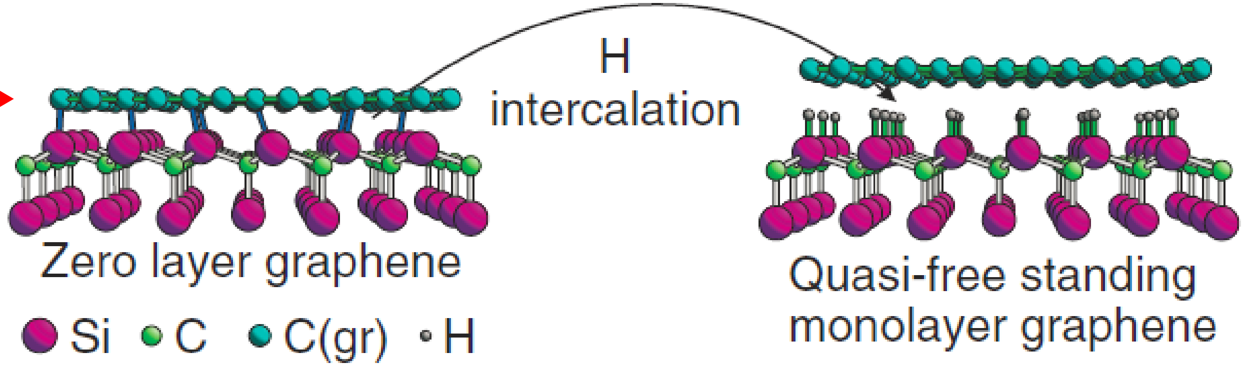
Highly-doped quasi-free-standing graphene

Buffer layer (not graphene)

Si-face of SiC



C. Riedl, et al., PRL 103, 246804 (2009)

Large scale monolayer graphene

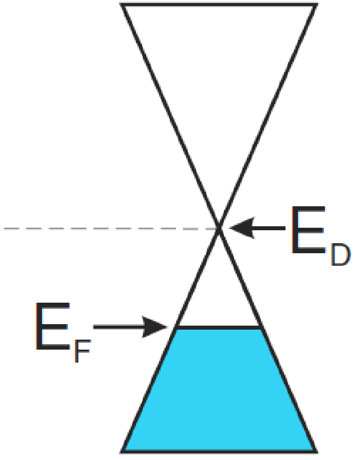
Highly p-doped graphene:

$$E_F \approx -300 \text{ meV}$$

Moderate mobility:

$$\mu \approx (2-3) \times 10^3 \, \text{cm}^2/(\text{V.s})$$

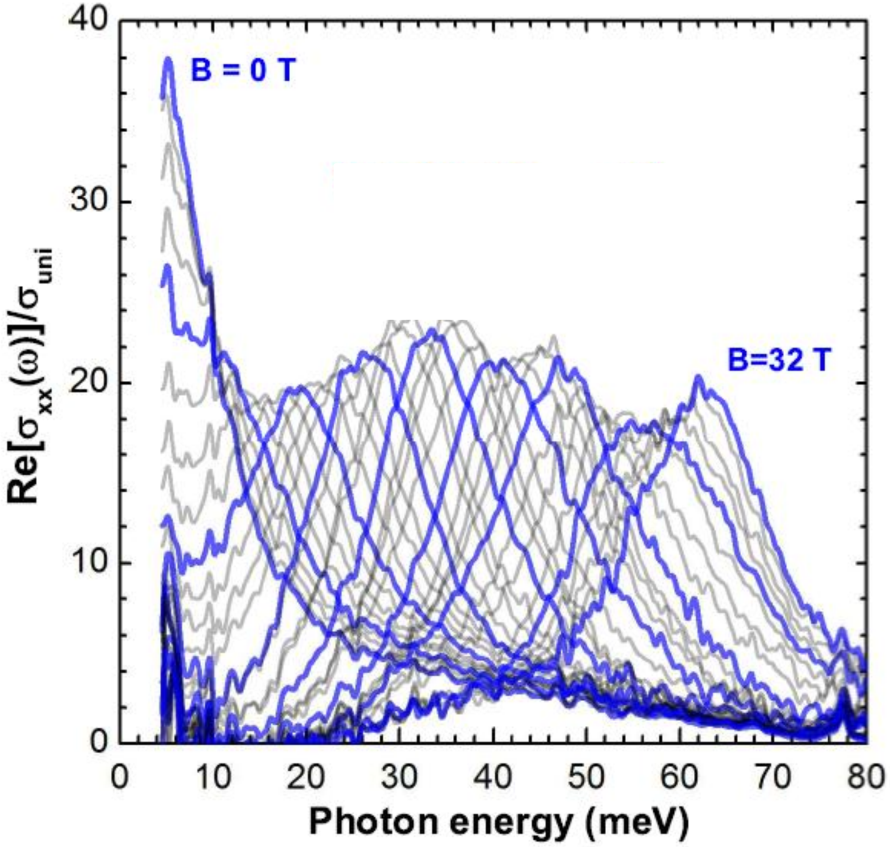
Samples: Th. Seyller, University Erlangen

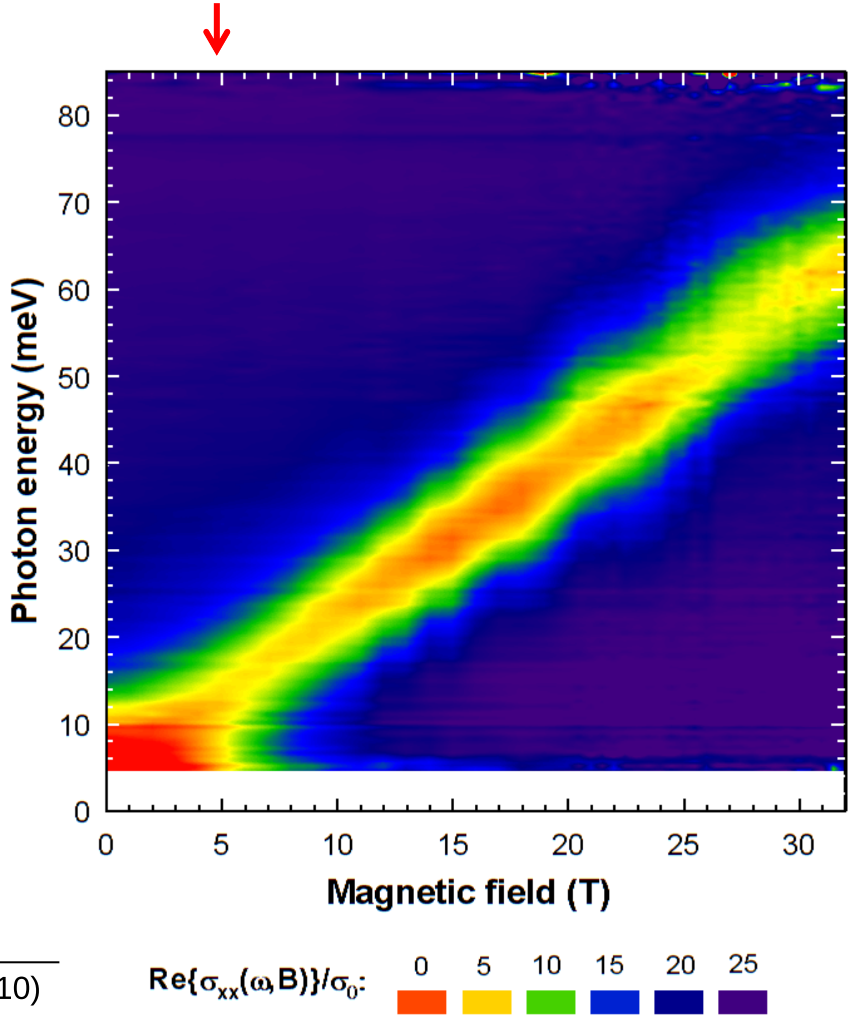


p-type

Cyclotron resonance in highly-doped quasi-free-standing graphene

$$\omega_c \tau = \mu B \sim 1$$





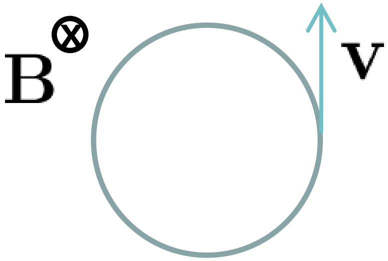
A. M. Witowski, M. Orlita et al., Phys. Rev. B 82, 165305 (2010)
M. Orlita et al., arXiv:1205.1118 (2012)

Cyclotron motion of massless Dirac fermions (classical regime)

Equation of motion for a charged particle in magnetic field (2D):

$$\frac{d\mathbf{p}}{dt} = e[\mathbf{v} \times \mathbf{B}]$$

Cyclotron motion at frequency:



$$\omega_c = \frac{eB}{\underbrace{(E/v_F^2)}}$$

Linear in magnetic field

Energy dependent

"Effective" effective mass
of massless particle (i.e., Einstein relation)

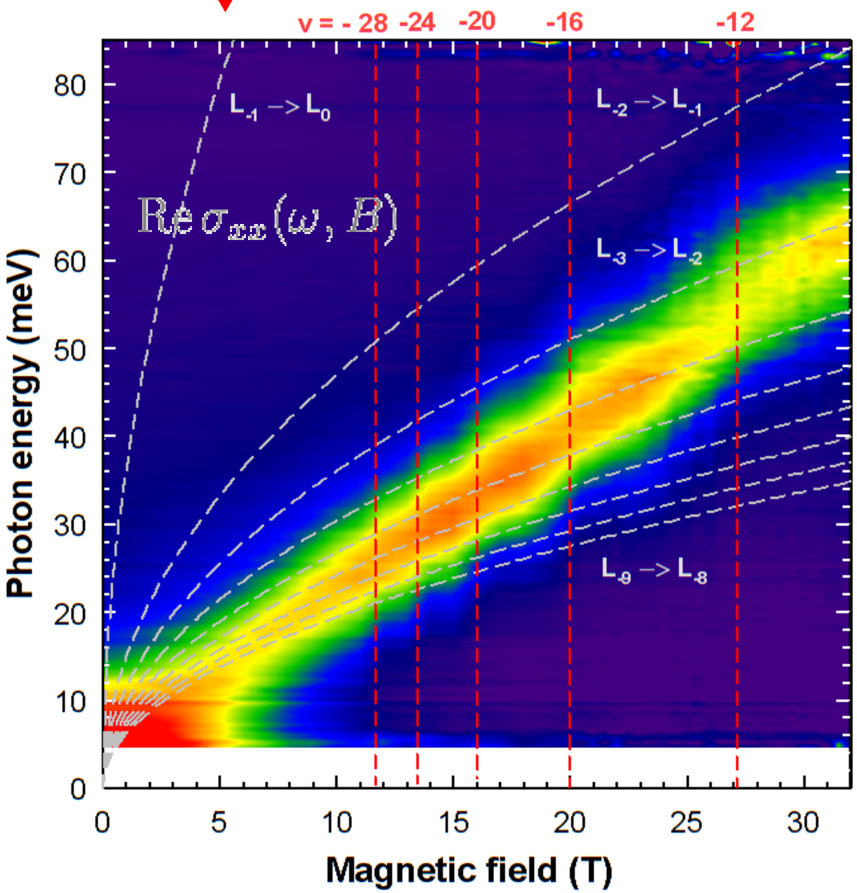
Comparison for massive particles (with a well-defined mass):

Linear in magnetic field, but energy independent
cyclotron motion at frequency:

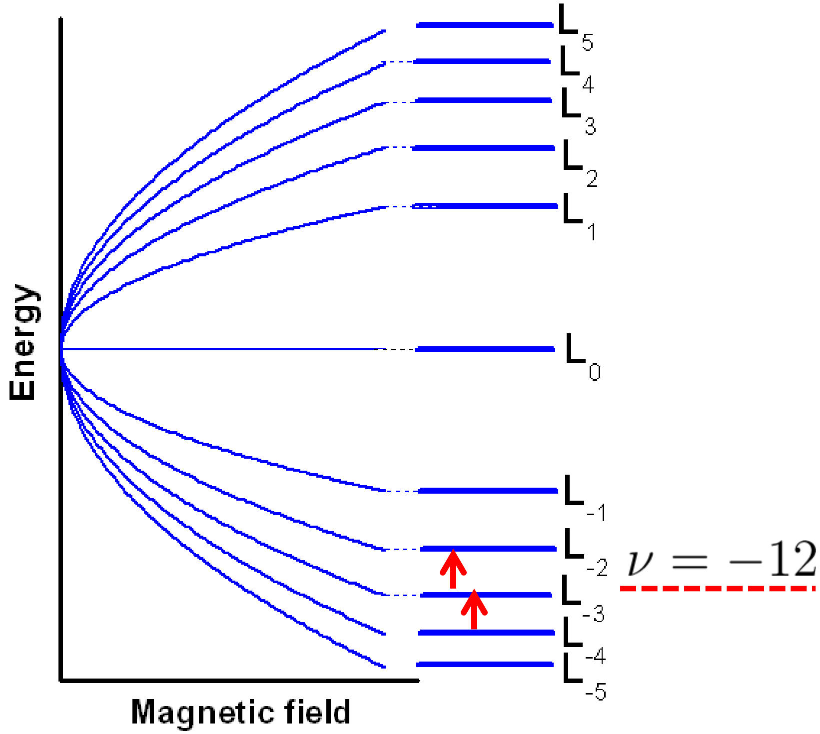
$$\omega_c = \frac{eB}{m}$$

Cyclotron resonance in highly-doped quasi-free-standing graphene

$$\omega_c \tau = \mu B \sim 1$$



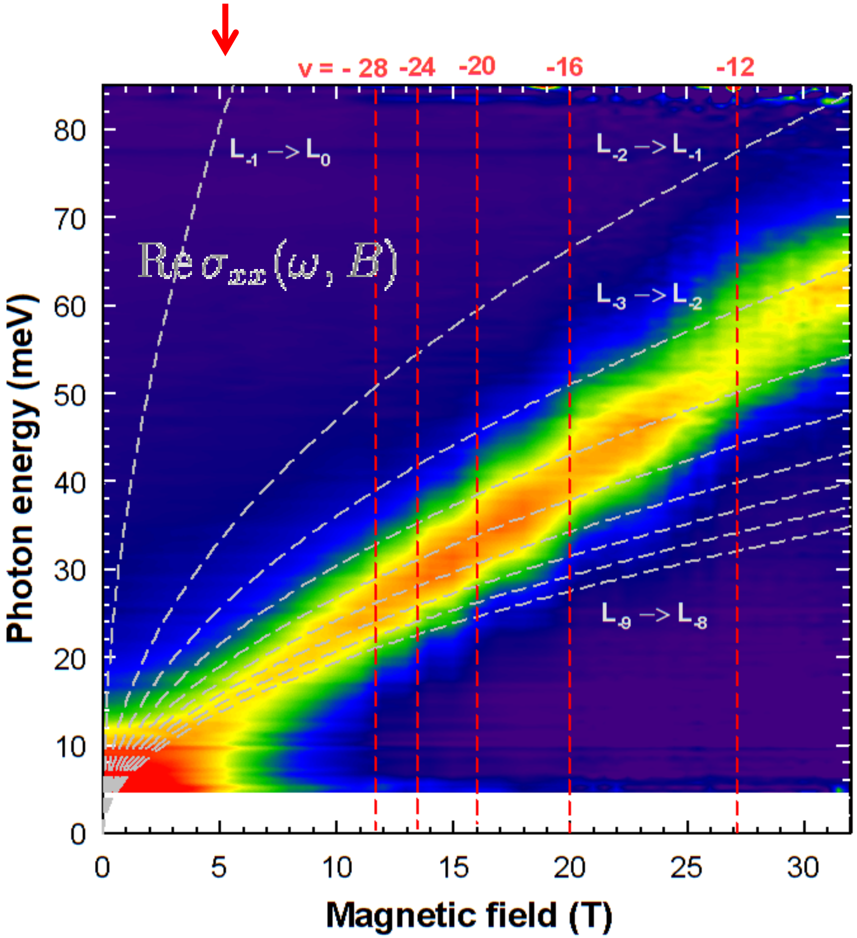
$$\omega_c = \frac{eB}{m} = v_F^2 \frac{eB}{E_F}$$



Crossover from classical to quantum regime

Cyclotron resonance in highly-doped quasi-free-standing graphene

$$\omega_c \tau = \mu B \sim 1$$



Crossover from classical to quantum regime

Sequence of filling factors

$$\nu = -4j, \;\; j = 12, 16, 20 \ldots$$

⇩

$$n = (7.9 \pm 0.2) \times 10^{12} \;\, \mathrm{cm}^{-2}$$

Carrier density & cyclotron mass

$$E_F = m_c v_F^2 \qquad E_F = v_F \hbar \sqrt{\pi n}$$

⇩

Fermi velocity:

$$v_F = (0.99 \pm 0.02) \times 10^6 \;\mathrm{m.s}^{-1}$$

Fermi level:

$$E_F = 325 \pm 5 \;\mathrm{meV}$$

M. Orlita et al., arXiv:1205.1118 (2012)

Total strength of intraband absorption = Drude weight

Drude weight in conventional (conducting) materials:

$$\mathcal{D} = \int_0^\infty \sigma_{\mathrm{intra}}(\omega)d\omega = \frac{\pi e^2}{2}\boxed{\frac{n}{m}}$$

Carrier density

Single-particle mass
(e.g., as measured in CR)

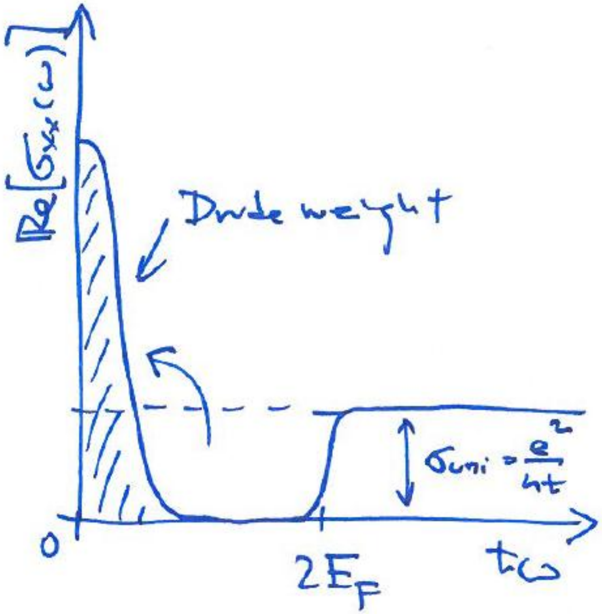
Justified for systems with Galilean invariance
(i.e., parabolic bands)

Drude weight in graphene?
Can we use an effective single-particle approach?

$$\mathcal{D} = \frac{\pi e^2}{2}\frac{n}{m} = \frac{e^2}{2\hbar}v_{\mathrm{F}}\sqrt{\pi|n|} = \frac{2\sigma_{\mathrm{uni}}|E_{\mathrm{F}}|}{\hbar}$$

Cyclotron mass
at the Fermi level:

$$mv_F^2 = E_F$$



Drude weight in graphene

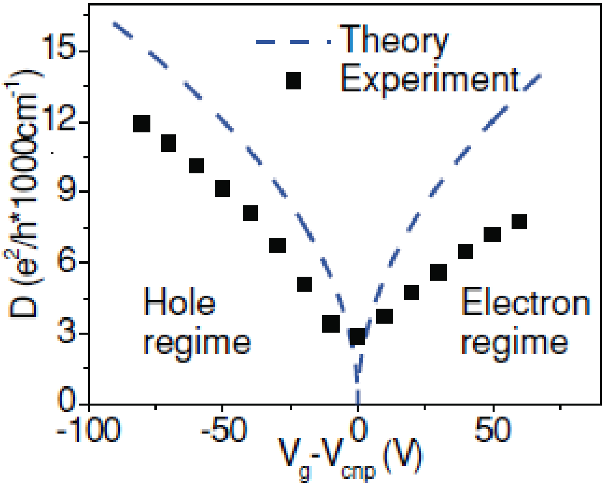
Single-particle approach justified theoretically only recently (for high carrier densitties only)**:**

S. H. Abedinpour et al., Phys. Rev. B 84, 045429 (2011)

$$\mathcal{D} = \frac{\pi e^2}{2}\frac{n}{m} = \frac{e^2}{2\hbar}\boxed{v_{\mathrm{F}}}\sqrt{\pi|n|} = \frac{2\sigma_{\mathrm{uni}}|E_{\mathrm{F}}|}{\hbar}$$

Drude weight follows the renormalization of the Fermi velocity

However, it is not supported by recent experiments…

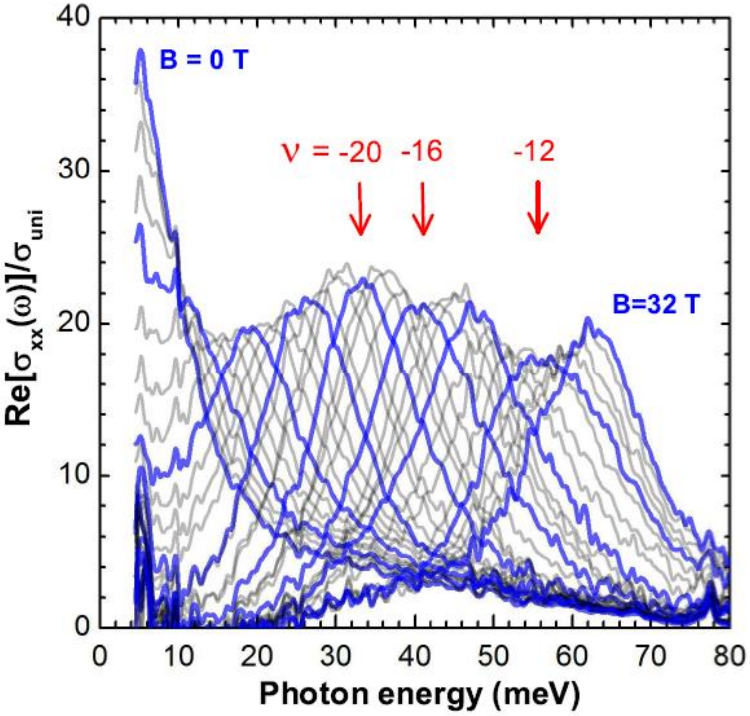


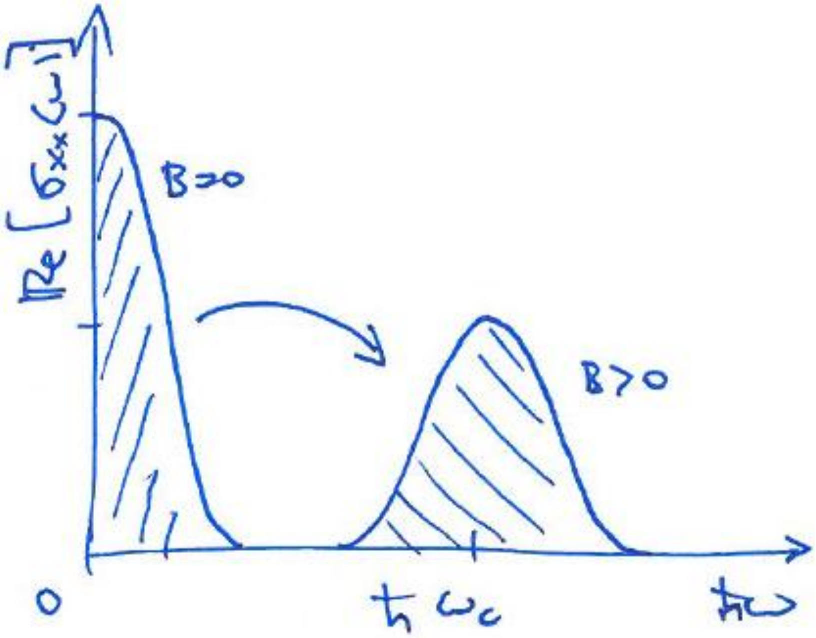
Significant supression of the Drude weight reported experimentally:

$$\mathcal{D}_{\mathrm{exp}} < \mathcal{D}$$

J. Horng et al., PRB 83, 165113 (2011)
H. Yan et al., ACS Nano 5, 9854 (2011)

Measuring Drude weight via strength of cyclotron resonance absorption





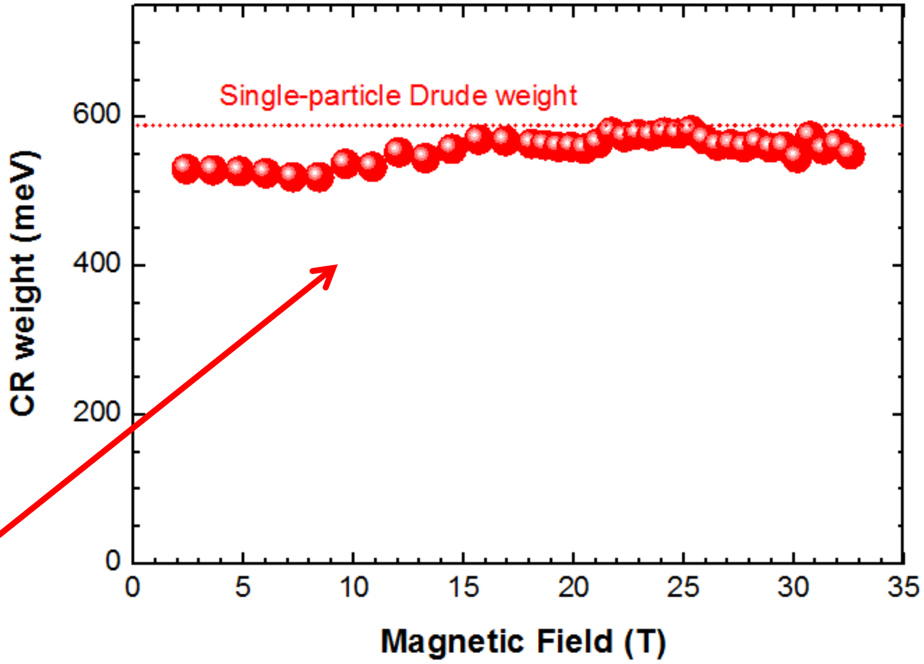
Single-particle picture valid ⇨ Drude weight fully transfered into the CR strength

Drude weight from cyclotron resonance strength

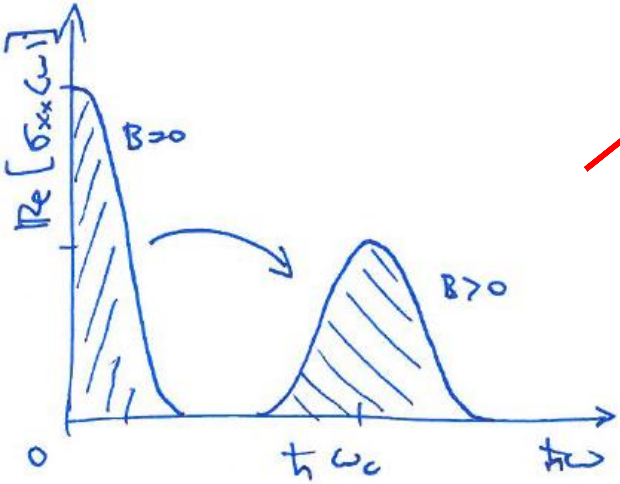
Quasi-free standing graphene = well-defined specimen
(carrier density, Fermi velocity, Fermi level…)

Expected single-particle Drude weight:

$$\mathcal{D} = \frac{e^2}{2\hbar} v_{\rm F} \sqrt{\pi|n|} = \frac{2\sigma_{\rm uni}|E_{\rm F}|}{\hbar}$$



Drude weight extracted directly from data:



No significant deviation from effective single particle picture

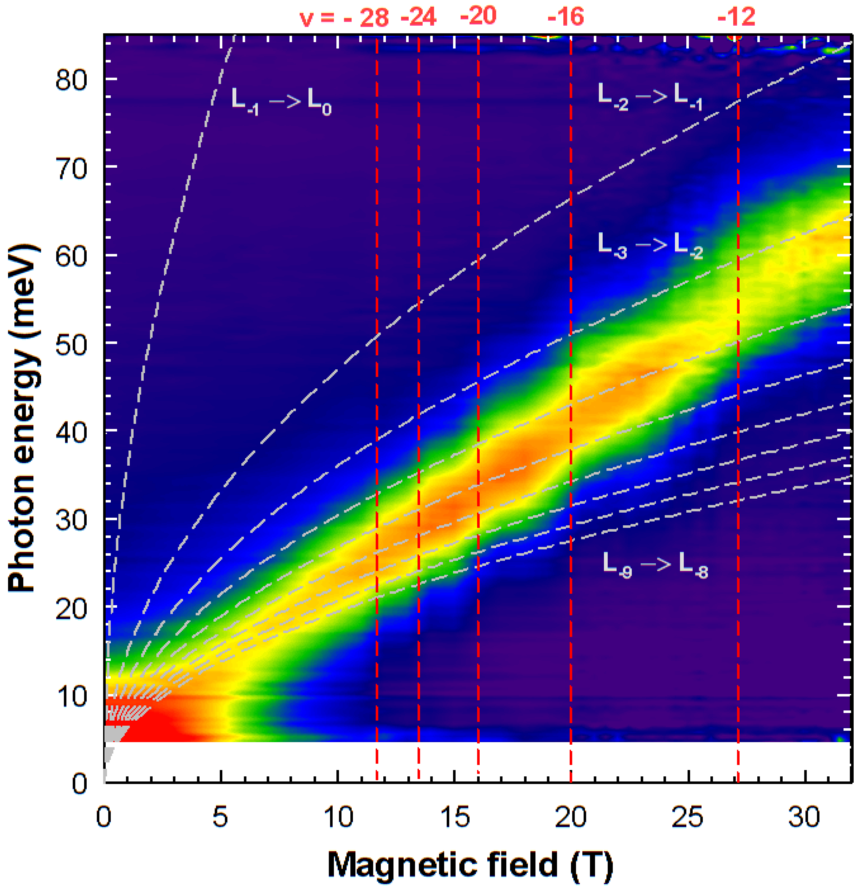
M. Orlita et al., arXiv:1205.1118 (2012)

Pisa, Italy

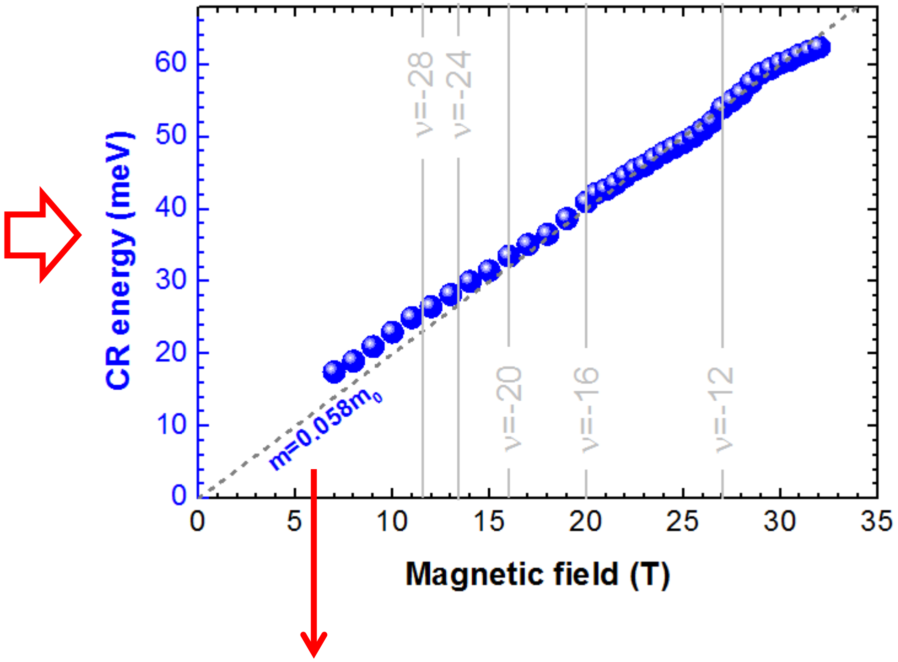
Collaboration with M. Polini

Cyclotron resonance in highly-doped quasi-free standing graphene: Analysis of low magnetic field data

Optical conductivity:



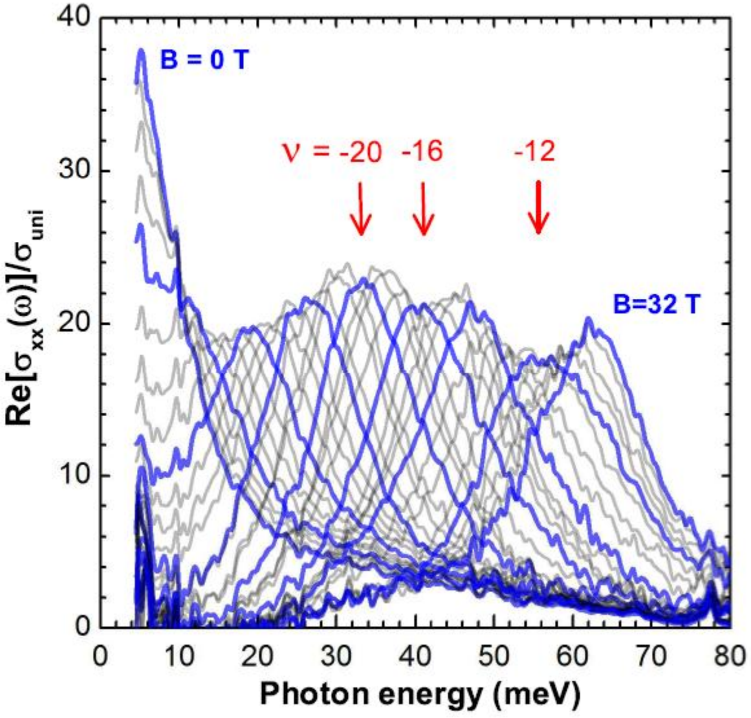
CR fan chart:

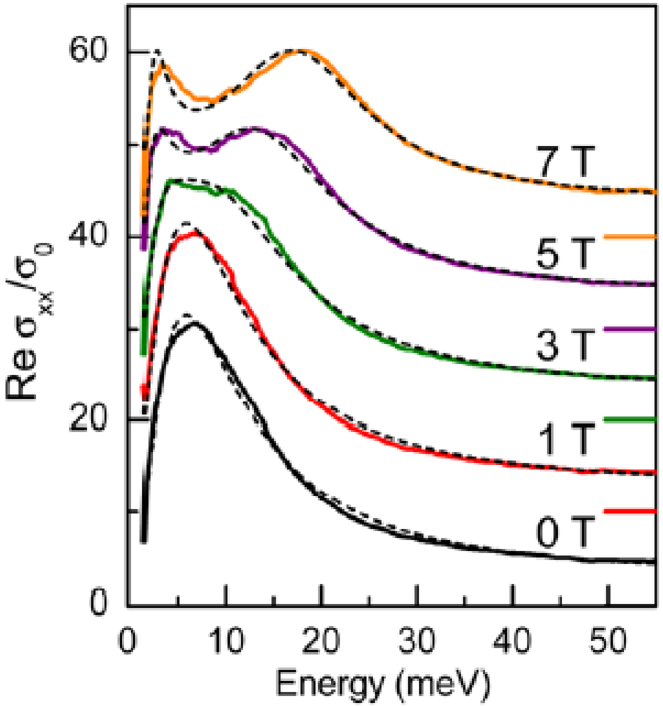


Low-field deviation of CR line from linearity in B

M. Orlita et al., arXiv:1205.1118 (2012)
I. Crassee, M. Orlita et al., Nano Lett. 12, 2470 (2012)

Optical conductivity of quasi-free-standing graphene at low frequencies





Drude peak (due to free carrier absorption) at non-zero energy

⇩

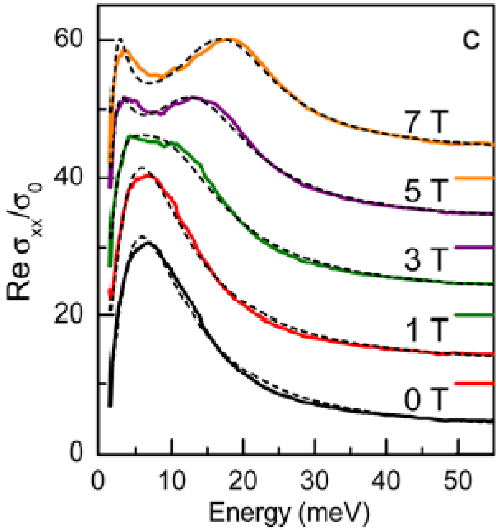
Signature of (confined) plasmons

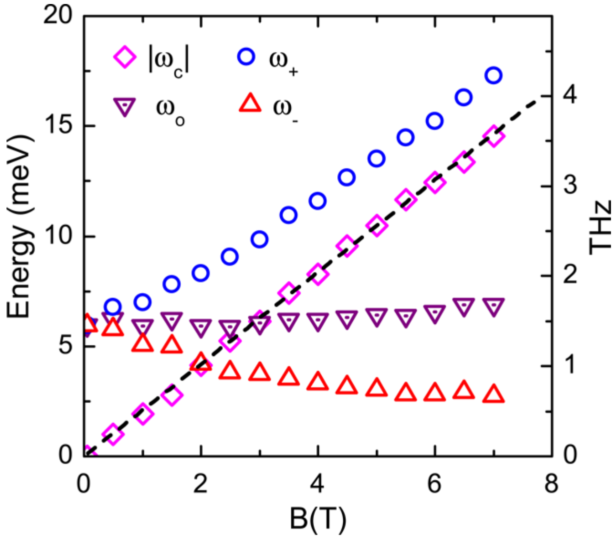
Collaboration with
I. Crassee & A. B. Kuzmenko

UNIVERSITÉ DE GENÈVE

I. Crassee, M. Orlita et al., Nano Lett. 12, 2470 (2012)

Plasmons in (unpatterned) quasi-free standing graphene?

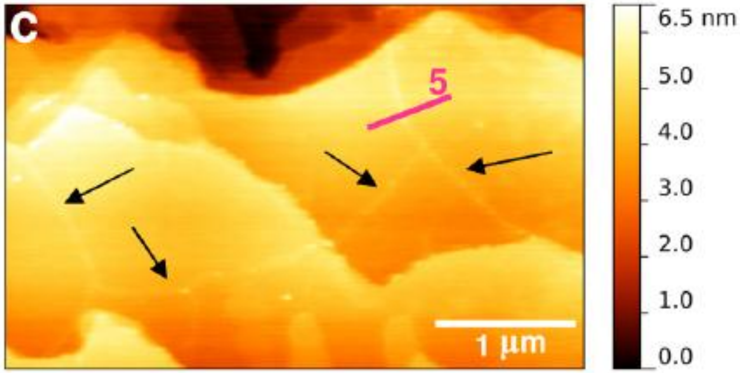




2D confinement of plasmons:

- **Characteristic scale: microns**
- **Dot-like geometry**
- **Weak (in-plane) anisotropy**

Typical AFM trace of quasi-free-standing graphene:

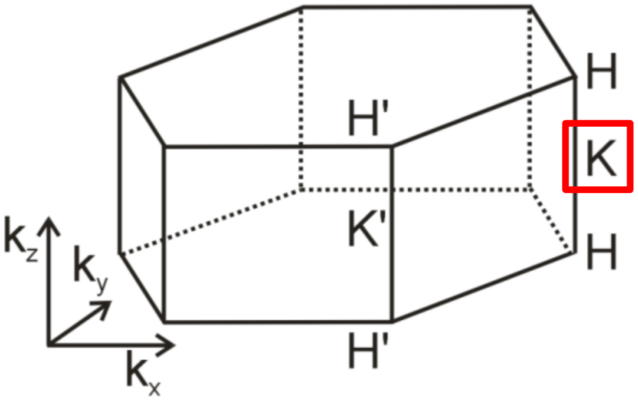


I. Crassee, M. Orlita et al, Nano Lett., 12, 2470 (2012)

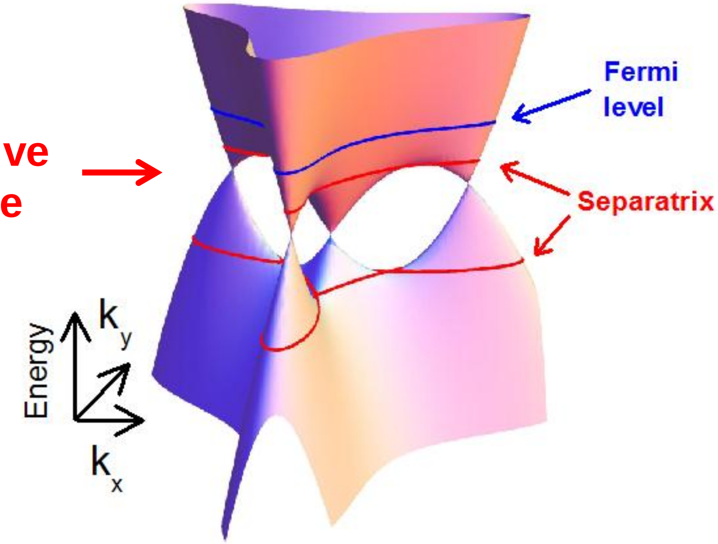
Origin of the 2D confinement:

Grain boundaries?
Substrate terraces?
….or?

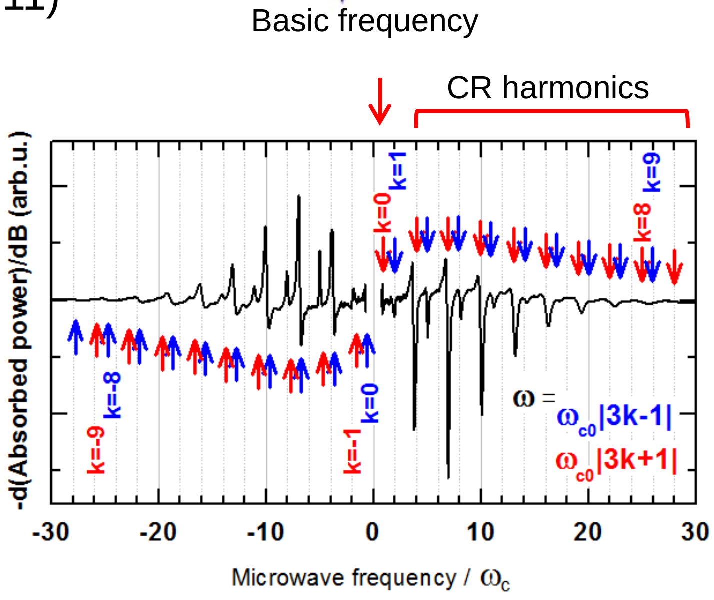
Lifshitz transition in bulk graphite



K point = effective bilayer graphene



Analog of the Lifshitz transition in bilayer graphene (Mayorov et al., Science 2011)

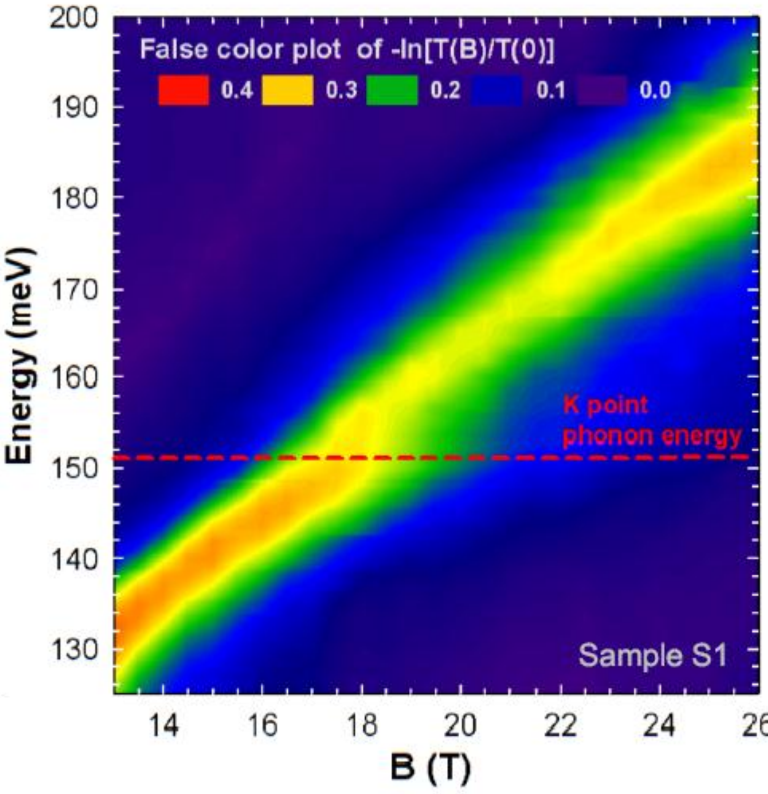


Graphite viewed as a system
in the vicinity of Lifshitz transition
via multimode CR response
in quasi-classical regime

Collaboration with D. M. Basko
LPMMC, CNRS, Grenoble

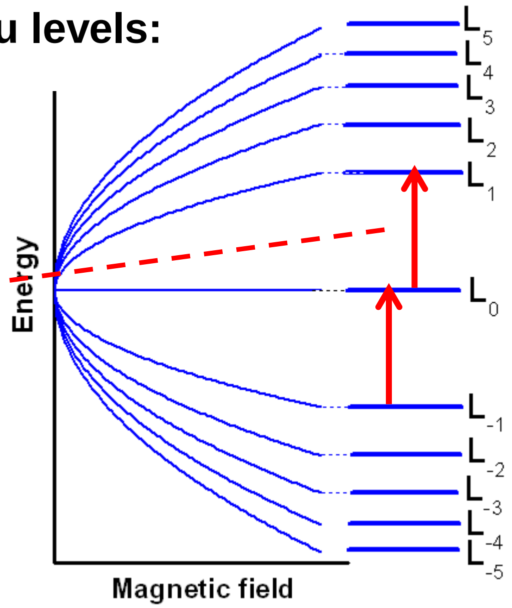
M. Orlita et al., PRL 108, 017602 (2012)

Interaction of K point phonons with cyclotron resonance



M. Orlita, et al. PRL 108, 247401 (2012)

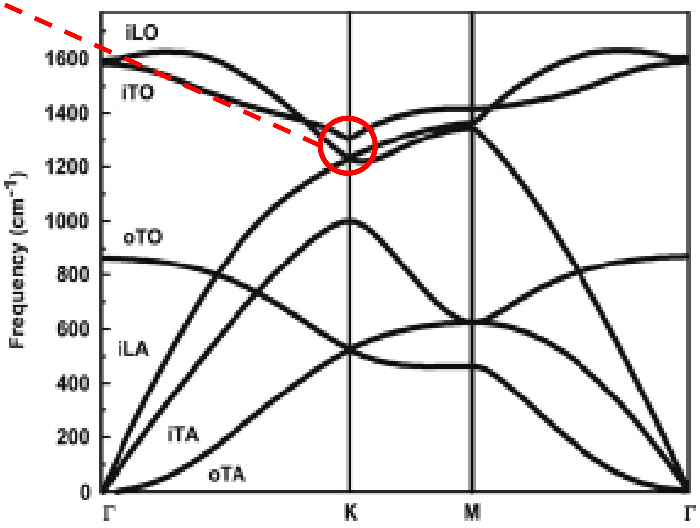
Landau levels:



Cyclotron resonance in quantum limit

Phonon dispersion:

K-point phonons responsible for 2D band in Raman



Collaboration with group of Steven G. Louie
University of California at Berkeley

Summary/Conclusions

Magneto-optics on graphene:

- (Magneto-)optical properties of graphene in both quantum and classical regimes
- Elastic and inelastic scattering of massless Dirac fermions
- Drude weight in graphene measured via strength of cyclotron resonance
- Evidence for “intrinsically” confined (magneto-)plasmons in quasi-free-standing graphene

Review article on optical properties of graphene:

M. Orlita and M. Potemski, Semicond. Sci. and Technol. 25, 063001 (2010) (arXiv:1004.2949)

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EC-EuroMagNETII under Contract No. 228043, Eurographene-EPIGRAT,
395/N-PICS-FR/2009/0, MSM0021620834, GACR P204/10/1020, MTKD-CT-2005-02967.

Acknowledgement: A.-L. Barra, R. Grill, D. K. Maude, W. Strupinski, P. Plochocka